Journal of Water Process Engineering 52 (2023) 103604

Contents lists available at ScienceDirect

Journal of Water Process Engineering

journal homepage: www.elsevier.com/locate/jwpe

Research advances in nuclear wastewater treatment using conventional and hybrid technologies: Towards sustainable wastewater reuse and recovery

Dana Kadadou [a,b,1], Eslam Ahmed Said [c,1], Rahaf Ajaj [c,*], Shadi W. Hasan [a,b,**]

[a] Center for Membranes and Advanced Water Technology (CMAT), Khalifa University of Science and Technology, PO Box 127788, Abu Dhabi, United Arab Emirates
[b] Department of Chemical Engineering, Khalifa University of Science and Technology, PO Box 127788, Abu Dhabi, United Arab Emirates
[c] Department of Environmental and Public Health, College of Health Sciences, Abu Dhabi University, Abu Dhabi, United Arab Emirates

ARTICLE INFO

Keywords:
Nuclear wastewater
Decontamination
Radioactive
Treatment
Recovery
Reuse

ABSTRACT

With the widespread implementation of nuclear energy, radioactive wastewater management is increasingly gaining research interest. As radioactive waste piles up, from nuclear applications among other industries, growing concerns on its impact on human health and the environment arise. This review comprehensively discusses the sources, classes, and occurrence of radionuclides in nuclear wastewater. An understanding of the nature of radioactive wastewater is vital for the selection of an appropriate management approach. Advances in conventional and hybrid technologies for the treatment of nuclear wastewater are also extensively reviewed and evaluated. Our review shows that adsorption and membranes are the most mature technologies in comparison with other chemical, biological, physical, and electrochemical processes. In fact, the combination of these techniques, in the form of adsorptive membranes, has also shown highly promising results as hybrid technologies. Lastly, this review highlights the overseen benefit of recovering valuable radionuclides from nuclear wastewater, which has not been addressed yet.

1. Introduction

The sustainable use of water, being the most valuable yet scarce resource on planet earth, is challenged by the growing population and increase in resource demand. With the emergence of contaminants in water bodies, increased industrialization continues to mark a decline on water quality. There is no question that the exploitation of natural resources on a global scale has an impact on urbanization and industrialization. An unforeseen type of pollution comes from radioactive waste. Nowadays, global energy demand is on the rise due to the growing population and increase in industrialization. Nuclear energy presents a viable option for electricity generation, and is well-known for its usage of low carbon outflow [1], unlike the traditional approach of utilizing fossil-based oils. In accordance, the dependence of countries on nuclear energy has been growing. In 2020, nuclear power plants (NPPs) accounted for 10.3 % of the world's total power generation, and by the end of 2021, the percentage had risen to 18.9 [2]. This trend is anticipated to continue in the coming years; thus, radioactive waste should be seriously considered and effectively managed.

Nuclear wastewater is produced in several applications such as

Abbreviations: NPP, Nuclear power plant; LRAW, Liquid radioactive waste; EW, Exempt waste; VSLW, Very short lived waste; VLLW, Very low level waste; LLW, Low level waste; ILW, Intermediate level waste; HLW, High level waste; IAEA-MEL, International atomic energy agency; HPGe, High purity germanium; BDC, Bamboo powder-derived biomass charcoal; HAP, Hydroxyapatite; ACM, Amidoxime cellulose microspheres; CMC, Carboxymethyl chitosan; LDH, Layered double hydroxide; MOF, Metal-organic frameworks; COF, Covalent organic frameworks; PAN, Polyacrylonitrile; MSP, Melamine styrene conjugated polymer; ACF, Amidoxime calixarene fiber; MVR, Mechanical vapor recompression; RO, Reverse osmosis; DTRO, Disk tubular reverse osmosis; HTGR, High-temperature gas-cooled reactor; FO, Forward osmosis; UF, Ultrafiltration; MD, Membrane distillation; DCMD, Direct contact membrane distillation; AGMD, Air gap membrane distillation; VMD, Vacuum membrane distillation; SGMD, Sweeping gas membrane distillation; NF, Nanofiltration; AL-FS, Active layer facing feed solution; AL-DS, Active layer facing draw solution; CDI, Capacitive deionization; FCDI, Flow electrode capacitive deionization; PAC, Porous activated carbon; ISESM, In situ electrochemical synthesis method; USM, Uranium-containing wastewater; CAPJ, Cold atmospheric plasma jet; BCF, Bioaccumulation factor.

* Corresponding author.
** Correspondence to: S. W. Hasan, Center for Membranes and Advanced Water Technology (CMAT), Khalifa University of Science and Technology, PO Box 127788, Abu Dhabi, United Arab Emirates.
E-mail addresses: rahaf.ajaj@adu.ac.ae (R. Ajaj), shadi.hasan@ku.ac.ae (S.W. Hasan).
[1] Dana Kadadou and Eslam Ahmed Said share first authorship.

https://doi.org/10.1016/j.jwpe.2023.103604
Received 25 November 2022; Received in revised form 13 February 2023; Accepted 21 February 2023
Available online 27 February 2023

nuclear energy, nuclear medicine, reprocessing facilities, and research reactors. Since large amounts of water are used to cool nuclear reactors, more liquid waste is generated than gaseous emissions in the nuclear industry. Similarly, other nuclear applications listed above tend to generate large amounts of nuclear wastewater. NPPs produce liquid radioactive waste (LRAW), which typically constitutes both soluble and insoluble radioactive components as well as non-radioactive materials. Radioactive waste generated by the nuclear industry is challenging to manage, and poses serious threats to human health and the environment. It not only induces radiation-related diseases, but can also cause biological mutations. Besides the health implication of radioactive waste, their unregulated discharge into the environment remains a concern for governments, researchers, and society. According to the International Atomic Energy Agency (IAEA), a typical large reactor produces around 25–30 t of used fuel per year. To date, it is estimated that 400,000 t of used fuel have been discharged from reactors across the globe, with only one-third having been processed [3].

The significance of radioactive waste management was evidenced in 2011, during the Fukushima Daiichi Nuclear Power Station disaster [4]. During the accident, various types of radioactive isotopes were released into water bodies. Upon the accident, radionuclides had been sequentially found in several water bodies. In such disastrous accidents, the marine environment is always the center of attention because most nuclear plants are generally located near the coast. Proper treatment and handling of nuclear wastewater can be accomplished through chemical, physical, electrochemical, or biological processes that either concentrate radionuclides in nuclear wastewater or separate them. At the time of the Fukushima Daiichi Nuclear Power Station accident, mature control strategies and treatment technologies were not readily available. However, research shows great advances in the treatment of radioactive wastewater from 2012 to 2022. Therefore, several studies have been reported to review single purification technologies to treat radioactive water [1–5]. Nevertheless, there are no recent comprehensive literature reviews on the treatment technologies of nuclear wastewater. This paper aims to bridge this existing gap in literature and shed the light on the importance of radioactive waste management. This review encompasses sources and classifications of radioactive waste, their environmental impact, occurrence and monitoring strategies, and extensively evaluates treatment approaches for handling radioactive wastewater. Recent advances in the treatment technologies, ion exchange/sorption, precipitation, evaporation, membrane separation, electrochemical processes, biosorption, and bioaccumulation are extensively reviewed. Finally, future research directions in treatment approaches and reuse applications are discussed.

2. Sources and classification of nuclear wastewater

Radioactive wastewater generated in the nuclear industry could come from NPPs, reprocessing facilities, research reactors, nuclear sector laundry, medical facilities, and labs [6]. This waste constitutes hazardous chemicals with high reactivity, low flash or ignition points, and the potential to produce explosive materials among other components when combined with other reagents. Generated radioactive wastewater also contains radioactive isotopes which are soluble in water. The properties and risks of all constituents in radioactive wastewater must therefore be carefully studied during decontamination, solidification, or disposal procedures. Generally speaking, a variety of radioactive materials are produced during NPP operations. The majority of these radioactive sources are created by the corrosion and nuclear fuel fission of activated byproducts. Liquid effluents from NPPs frequently release a small quantity of these radioactive sources into the environment during normal operation. The majority of the time, these radioactive wastes originate from a variety of sources, including corrosion of active substances from pipes, valves, pumps, and other machinery. Previously, radioactive wastewater was not handled and treated in the most appropriate ways. As a result, radioactive wastewater with unidentified components has accumulated with time [6]. Moreover, crucial components used to slow down rapidly-moving neutrons in nuclear reactors are moderators. Each fission of uranium-235 produces rapidly-moving neutrons to start another reaction, which can only be slowed down by the addition of moderators. Among the most commonly utilized moderators is water. Kinetic energy released during the fission of uranium is carried by the fast-moving byproducts of the fission process. An enormous amount of this energy is removed by the fast neutrons, which is then dissipated in their collisions with the atoms and molecules in the reactor core. In many reactors, water is used as a moderator to absorb this energy, slowing down the neutrons. Since water is used in the majority of nuclear reactors, LRAW is consequently generated, and this is believed to be the main source of radioactive wastewater in the nuclear industry [7].

According to Burcl (2013), the primary source of LRAW is nuclear energy production, and the main isotopes in LRAW are Co-58, Co-60, Mn-54, Fe-59, Cr-51, and Sb-124, which are produced by the neutron activation of pipes and contour corrosion products. The remaining isotopes are Sr-90, Y-90, Cs-134, and Cs-137, which arise from untight heat-emitting materials [8]. In NPPs, the quality of generated LRAW mainly depends on whether spent fuel is reprocessed or thrown away [9]. Moreover, the LRAW's origin typically reveals its key traits and serves as the primary source of data for categorization, classification, analysis, and decision-making regarding the various eradication methods. There are numerous ways to describe and group LRAW. The main characteristics include chemical composition, aggregate condition, mechanical qualities, and radiological properties, such as the activity concentration, type, and physical parameters of the radionuclides present in the waste [9]. The classifications and discharge methods of radioactive wastewater are visually demonstrated in Fig. 1. Wastes that meet the requirements for clearance, exemption, or exclusion don't need regulatory control and are called exempt wastes (EW). In that case, the management and disposal of EW are not subject to any governmental rules [9,10]. The regulatory authority has approved the short-term storage, up to a few years, of LRAW containing radionuclides with very short half-lives while the radionuclides degrade. After that, it is no longer under regulatory scrutiny. The majority of the time, “monoisotopic” LRAW from institutional uses of radionuclides, particularly in medicine and research, falls into this category. To ensure safety during decay, the right storage conditions must be established. These wastes are referred to as very short lived waste (VSLW). Wastes that fall under the very low-level waste (VLLW) category are those whose activity concentration is slightly higher than that accepted for EW, but VLLW does not require high-level isolation and confinement. With little regulatory oversight, it is possible to dispose of VLLW at landfill-style facilities, like those used, for example, for non-radioactive hazardous waste.

When nuclear facilities are shut down, enormous amounts of contaminated soil, debris, concrete, thermal insulation, and other materials are produced. Long-lived radionuclides are present in very small amounts in both VLLW and low-level waste (LLW); however, LLW is distinguished with radiation concentrations that are significantly above clearance criteria. The main sources of LLW are NPP operations and waste, which can take on a variety of physical forms and have different chemical and radiochemical compositions. Numerous radionuclides (fission products, activated products, etc.) exist in a variety of activity concentrations. This waste needs to be isolated in order to be processed, stored, and contained for up to a few hundred years (300 years is typically used because that is ten times the half-life of Cs-137). It can be disposed of in locations made to be close to the surface after being properly processed and contained [9,10]. Furthermore, waste designated as intermediate level waste (ILW) is waste that has a higher radioactivity concentration or a higher concentration of long-lived radionuclides than waste classified under the previous LLW category. For example, long-lived fission products and alpha-emitting radionuclides might be present in ILW and might not degrade to a concentration of activity suitable for disposal close to the surface within the time frame

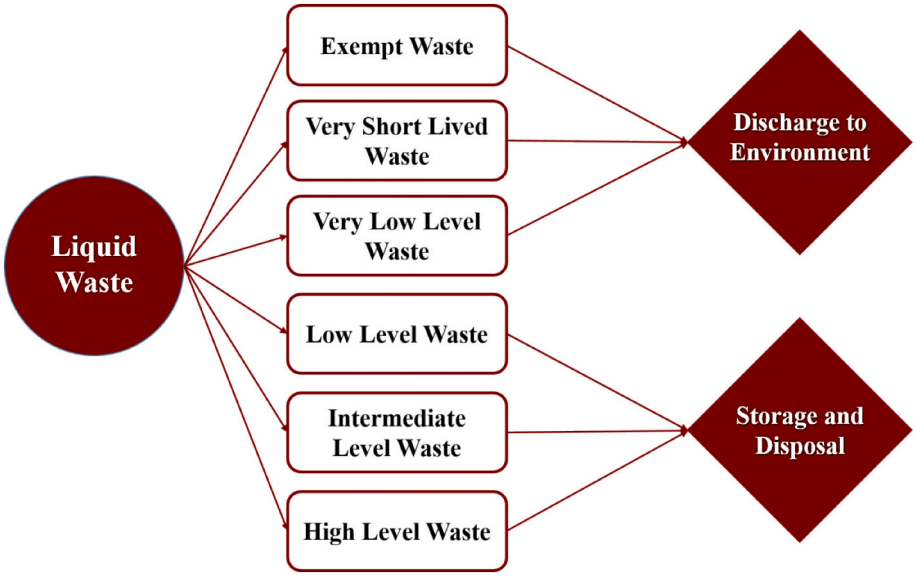

Fig. 1. Classification of radioactive liquid waste, adapted from [9].

for which institutional controls might be relied upon. While being handled, stored, and disposed of, ILW requires little to no provision for heat dissipation [9]. It is debatable whether to distinguish between LLW and ILW using minor long-lived radionuclides or the activity concentration of bulk radionuclides. The upper activity concentration limits for the ILW are also a subject of disagreement. The reprocessing of used fuel is a significant additional source of ILW at NPPs, which is typically produced as a byproduct of the main waste treatment (i.e., concentration). In comparison to the disposal of LLW at shallow depths, the disposal of ILW at depths of tens to hundreds of meters in underground repositories requires a higher level of containment and isolation [10].

When deciding where and how to dispose of high-level waste (HLW), it is important to keep in mind that it has radionuclide concentrations and activity levels that can generate a lot of heat through radioactive decay. In some circumstances, HLW needs to be handled and stored with additional cooling. Highly active reactor components are common examples of HLW made at NPPs. However, the main source of HLW is the reprocessing of spent nuclear fuel [9]. Most experts concur that storing HLW in stable, deep geological formations that are typically several hundred meters below the surface is the best way to dispose of it [10].

3. Occurrence and monitoring of radionuclides

The occurrence of radionuclides in wastewater is expected due to improper handling practices which do not follow those described in Section 2. In brief, all medical and NPP outputs lead to the introduction of radioactive effluents into water bodies. In a hospital setting, radiopharmaceuticals may be administered intravenously, orally, or inhaled, depending on the test. Depending on the mode of administration, the excretion pathway changes. Urine is one of the excretion mechanisms, and it frequently allows radionuclides to enter the sewage system. If a diagnostic dose is administered, this might occur immediately through patient discharges. Alternatively, it might happen after radioactively decaying in hospital waste storage tanks. In NPPs, very minute quantities of particular radionuclides are released through liquid and aerosol discharges and may end up in the environment [11]. Several research articles have detected the presence of liquid radionuclides in effluents of several applications in the nuclear industry in different regions of the world, some of which are listed in Table 1.

As a response to the occurrence of radionuclides in processes' effluents, and their anticipated discharge into water bodies, regular monitoring should be ensured to control the radioactivity being released.

Table 1
Summary of released radionuclides from nuclear power plants, nuclear reprocessing plants, and nuclear medicine in different regions of the world.

Nuclear industry	Region of study	Released radionuclides	References
Nuclear power plants	USA	Iron (55), Cobalt (58, 60), Cesium (134, 137), Chromium (51), Manganese (54), Zirconium (95), Niobium (95), Iodine (131, 133, 135), Hydrogen (3), Krypton (85, 85-m, 87, 88), Xenon (131, 133, 133m, 135, 135-m)	[12]
	France	Hydrogen (3), Carbon (14), Manganese (54), Cobalt (58, 60), Nickel (63), Silver (110-m), Antimony (124), Iodine (131, 133, 135), Cesium (134, 137)	[13]
	South Korea	Carbon (14), Dissolved noble gas (Xe-133), Particulate Antimony (122, 124, 125), Cesium (134, 137), Cobalt (56, 57, 58, 60), Iron (55), Manganese (54), Niobium (95, 97), Strontium (89, 90, 92), Zirconium (95), Iodine (131,133)	[13]
Nuclear reprocessing plants	USA	Natural Uranium, Radium (226), Thorium (230)	[12]
	France	Hydrogen (3), Carbon (14), Particulate Antimony (125), Cesium (134, 137), Cobalt (56, 57, 58, 60), Iron (55), Manganese (54), Zinc (65), Niobium (95, 97), Strontium (89, 90, 92), Zirconium (95), Technetium (99), Iodine (131, 133), Cerium (144), Europium (154), Natural Uranium, Plutonium (238, 239, 241), Americium (241), Curium (244).	[14]
	Australia	Hydrogen (3), Plutonium isotopes, Cesium (137), Strontium (90), Ruthenium (106), and Iodine (129)	[15]
Nuclear medicine	Spain	Gallium (67), Technetium (99-m), Lanthanide (111), Iodine (131), Carbon (14), Hydrogen (3)	[16]
	Italy	Fluorine (18), Technetium (99-m), Lanthanide (111), Iodine (123), Iodine (131)	[17]

Routine wastewater samples should be analyzed to determine their radionuclide activity concentrations and radiation fields. Establishing a baseline quality, identifying environmental trends, differences, new environmental problems, and assessing the degree to which environmental objectives are being met are all examples of environmental monitoring assessments [18]. Routine monitoring, emergency preparedness, and emergency monitoring are the three different types of monitoring programs. Some detection techniques have already been proposed and utilized. For example, in situ gamma-energy spectroscopy was first used in seawater around 1950 to control radiation levels. Presently, some countries have automatic online measurements of ocean gamma-energy spectra. In order to expand the monitoring range to various marine environments, the Marine Environment Laboratory of the IAEA-MEL developed a dual-probe marine in situ gamma spectrometer using a High Purity Germanium (HPGe) semiconductor with high energy resolution and a NaI(Tl) scintillator with high detection efficiency [19]. The tool was employed for environmental radioactivity surveys at a nuclear waste disposal site in the Kara Sea and a seabed sediment gamma-ray survey in the Irish Sea close to the Sellafield nuclear fuel reprocessing plant in the UK to ascertain the distribution and inventory of Cs-137 and I-131. NaI(Tl) detectors are frequently used in seawater in-situ gamma spectrometers due to their low cost, superior detection efficiency, and wide operating temperature range, despite lacking energy resolution. Moreover, Guoxiu et al. (2017) use an HPGe detector which has a strong energy resolution, improving peak separation and radionuclide detection [20]. However, the HPGe system is unreliable as it consumes more power than low and medium-resolution systems and cannot operate continuously for long durations. Therefore, any detector with a high detection efficiency and sufficient energy resolution at room temperature would present an ideal device for in situ gamma-ray spectrometry and long-term monitoring in seawater.

4. Treatment technologies used for nuclear wastewater

As previously mentioned, the world's dependence on nuclear energy is escalating. It is therefore realized that with time, the dependence on

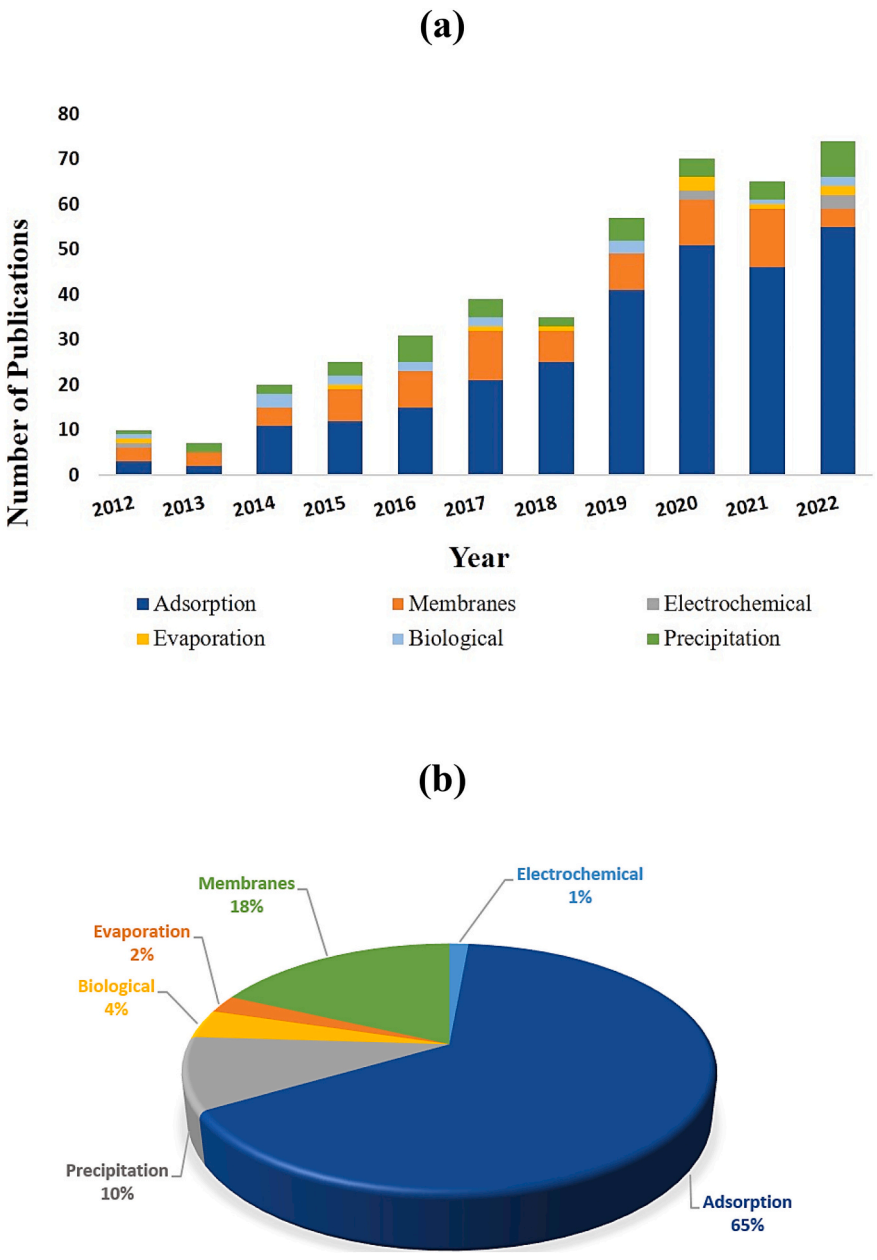

Fig. 2. (a) Number of publications on radioactive wastewater treatment utilizing adsorption, membranes, electrochemical methods, evaporation systems, biological processes, and precipitation indexed by Scopus from 2012 to 2022. (b) Distribution of studies focusing on adsorption, membranes, electrochemical methods, evaporation systems, biological processes, and precipitation. Source: Scopus index and Sciencedirect with keywords: "radioactive wastewater" AND "adsorption" OR "membrane" OR "electrochemical" OR "evaporation" OR "biosorption" OR "bioaccumulation" OR "precipitation" (extracted October 2022).

nuclear energy is expected to increase. Accordingly, the growing need for the treatment of radioactive wastewater generated in NPPs is anticipated. Indeed, Fig. 2(a) confirms this need with the annually increasing number of published articles in the field of radioactive wastewater treatment. This emerging field of research indicates the urge to reuse treated nuclear wastewater or perhaps recovered radionuclides, which is discussed in Section 7. Thus far, various chemical, physical, biological, and electrochemical methods have been explored for nuclear wastewater treatment, with adsorption and membrane separation being the most heavily researched, as depicted in Fig. 2(b). The subsequent sections critically review and analyze recently reported treatment technologies. Apart from these conventional treatment technologies, thermal plasma treatment (TPT) has also been explored by some researchers for radioactive waste management [21,22]. Despite being highly promising as a volumetric reduction, vitrification, gasification, and radionuclide retention modeling method, as well as a thermal processing system, TPT has yet to be studied in more depth for its applications in nuclear waste management [23].

4.1. Chemical processes

4.1.1. Ion exchange/sorption

4.1.1.1. Natural adsorbents. Recently, great research advances have been seen in the development of synthetic or engineered adsorbents. Despite their high adsorption capacity, these adsorbents remain costly solutions to the treatment of wastewater. Thus, renewable natural resource materials such as clays, minerals, and biopolymers are favored. Several studies suggest that natural adsorbents can be utilized for the adsorption of radionuclides. Sun et al. (2022) studied bamboo powder-derived biomass charcoal (BDC) as an adsorbent of uranium [24]. BDC was composited with MoS_2 and modified with phytic acid to enhance its adsorptive performance. Evidently, kinetics data was in line with the pseudo-second-order model and thus, the removal mechanism was confirmed to be chemical adsorption. Hydrogen plasma treatment was also applied to increase sulfur vacancies, enhancing the adsorption process. A naturally abundant mineral, hydroxyapatite (HAP), was investigated for its adsorptive performance towards uranium. HAP is an environmentally friendly, biodegradable, and low-cost material. The prepared HAP aerogel exhibited a continuous porous structure which led to a high adsorption capacity of uranium due to the presence of a high surface area. A 99.4 % uranium removal efficiency was achieved within only 10 min of contact time [25].

Cellulose, a natural biopolymer, has also been explored as a biodegradable material for the adsorption of uranium. In addition to being readily available, cellulose has rich reactive side groups such as hydroxyl groups. As is the case with most natural adsorbents, modifications to cellulose are needed in order to enhance its selectivity towards uranium. In a study by Wu et al. (2021), high grafting densities of amidoxime groups on cellulose were achieved to form highly adsorbing amidoxime cellulose microspheres (ACM) [26]. The prepared ACM could selectively decontaminate 390 mg g^{-1} uranium from simulated wastewater. Similarly, a natural *co*-polymer, chitosan, was used by Ding et al. (2022) to prepare carboxymethyl chitosan (CMC) using a new crosslinker, iminodi-acetic acid [27]. CMC possesses high water solubility and functional groups which are electrostatic providers. Such properties are ideal in metal ion adsorption. Moreover, bentonite, a naturally occurring clay, has been used for the adsorption of radionuclides. Since it has a high affinity towards cationic pollutants, bentonite is a very poor adsorbent of anionic compounds. As such, it has been previously used for the adsorption of positively charged radionuclides in water. To enhance its performance towards anionic compounds, researchers such as Yang et al. (2021) have investigated modifications by organic cations [28]. HDPy-bent is composed of bentonite modified with hexadecyl-pyridinium. In their study, the authors report great adsorption capacity towards Tc(99-m). It is analyzed that modifying bentonite with organic cations provides an abundance of adsorption sites for anionic compounds such as $^{99}TcO_4^-$. It is worth mentioning that synthetic clays have also been researched. For example, layered double hydroxides (LDH) have been explored for the adsorption of cesium and uranium [29,30].

4.1.1.2. Engineered adsorbents. It should be acknowledged that achieving high adsorption capacities with natural adsorbents is highly challenging, as suggested by prior research [27,30,31]. Therefore, most early studies as well as current work focus on developing synthetic or engineered adsorbents for the decontamination of radioactive wastewater. Previously, radionuclide adsorption studies were conducted using carbonaceous, polymeric, oxidic, and zeolite molecular sieves adsorbents among others. Studies utilizing metal oxides adsorbents such as titanium oxide are well documented [32–35]. It is also well-acknowledged that they possess remarkable uranium adsorption capability since a very long time ago [34]. For example, recently, research has shown promising evidence for titanium oxide-silver oxide composite as a radionuclide adsorbent. Mahmoud et al. (2018) developed nano titanium oxide-coated-silver oxide for the adsorption of zinc and cobalt from radioactive wastewater [35]. However, the authors give some information on its limitations, comprising its poor mechanical strength which limited its further exploration by other researcher groups. Industrial applications require sufficient adsorbent mechanical stability to withstand high bed resistance encountered in columns. Thus, Tang et al. (2021) proposed supporting highly performing adsorbents on polymer matrices. In their experiment, the authors prepared hydrous titanium oxide-immobilized collagen fibers (HTO/CFs). The performance of the synthesized adsorbent was tested in both batch and column adsorption setups. Results indicate an adsorption capacity of 1.379 mmol g^{-1} [34], indicating that titanium oxide composites can perform as efficiently with support layers. Unlike metal oxides, metal sulfides have just recently emerged as a new class of adsorbents with highly promising adsorption kinetics, selectivity, and capacity. Thus, there is a wide choice of metal sulfide adsorbents available in the literature [36–40]. Zhang et al. (2018) have developed metal sulfides KZTS and KZTS-NS for the removal of strontium and cobalt, respectively [39,40]. In their latter study investigating the performance of KZTS-NS, results indicate a significantly higher adsorption capacity than that of KZTS, despite exhibiting slow adsorption kinetics.

A closer look to the research on carbonaceous adsorbents, however, reveals a number of gaps and shortcomings. Research on carbonaceous adsorbents has been limited due to their small pore sizes and pore volumes. In the light of the recently reported studies on porous carbon-based adsorbents, it is conceived that they possess unique properties as compared to other types of radionuclide adsorbents. MXenes, for instance, is a newly emerging two-dimensional carbonaceous material that is produced by the exfoliation or etching of the A layer from the MAX phase. Enhancing their selectivity in adsorption has been seen by proper functionalization or by developing porous MXene gels which are three-dimensional in structure. Liu et al. (2022) developed magnetic amidoxime-functionalized MXenes for the selective removal of uranium and thorium. After conducting adsorption experiments, the authors confirm the suitability of the as-synthesized MXene material for the decontamination of radioactive ions [41]. Jun et al. (2020) also functionalized MXene and developed $Ti_3C_2T_x$ to remove the radionuclide cesium from wastewater [42]. In addition to its excellent adsorption capacity, the authors confirmed its reusability for at least 4 cycles, making it a practical and economical adsorbent. Other research groups have used porous carbon which is prepared by several methods such as pyrolyzing organic precursors and metal-organic frameworks (MOFs)-derived techniques. Sun et al. (2021) synthesized MOF-derived phosphorylated porous carbons, which showed high adsorption capability towards uranium with a capacity of 496.5 mg g^{-1} [43]. Alternatively, to

obtain a highly porous structure, covalent organic frameworks (COFs) are used and conjugated with carbon [44].

Furthermore, a number of authors have realized the unique advantages of silica-based materials due to their excellent radiation stability, which is of high significance in the application of nuclear wastewater treatment. Mesoporous silica materials such as SBA-15 and MCM-41 present themselves as ideal adsorbents due to their simple transformation into stable waste forms. For instance, the SBA-15 adsorbent developed by Ding et al. (2020) for uranium adsorption can be transformed into cristobalite ceramic waste of high durability [45]. Uranium, in nature, possesses a high affinity towards organic ligands and thus, waste management is a vital criterion to consider from an environmental point of view. The modification of another mesoporous material, MCM-4, has been explored by several researchers. Aluminum doping, tri-n-butyl phosphate (TBP) grafting, and Fe_3O_4 functionalization of MCM-4 were explored by different research groups [46–48]. Among the synthesized adsorbents, MCM-Zr-TBP performed better than Fe_3O_4@MCM-41-PDA/OA in terms of removal capacity and rate in adsorbing europium. This may be attributed to the grafted alkyl phosphate, TBP, which offers good adsorption, selectivity, and high extraction capabilities.

Polymeric adsorbents were also modified to treat radioactive wastewater. A novel polyamine-modified polyacrylonitrile-based fiber (PAN_{PA}) exhibited outstanding adsorption capability, 459.27 mg g^{-1}, towards uranium in the presence of F^- in high concentration [49], while two aminotriazole isomers modified microcrystalline cellulose microspheres exhibited good adsorption capacity for uranium at a relatively slow rate [50]. Additionally, melamine styrene conjugated polymer (MSP) and Prussian blue (PB)-immobilized adsorbent (PP-g-PAA-PB) were prepared for the decontamination of cesium [51,52]. Both MSP and PP-g-PAA-PB had similar adsorption capacities and reached equilibrium within 60 min. Mesoporous graphite carbon nitride (*mpg*-C_3N_4) was synthesized for the adsorption of thorium and showed high adsorption performance in pH 4.0, making it an ideal candidate for real nuclear wastewater which is often slightly acidic in nature [53]. Moreover, amidoxime calixarene fiber (ACF), fluorine-9-bisphenol (PHCP-2), and amidoximation ethylene-acrylic acid copolymer balls (EAA-AO) were developed for the adsorption of uranium [54–56]. Among these novel adsorbents, PHCP-2 exhibited the highest adsorption of uranium and EAA-AO showed ideal performance under acidic conditions (pH 4.0). Table 2 summarizes the recent natural and synthetic adsorbents utilized for radioactive wastewater treatment.

4.1.2. Precipitation

A series of recent studies has indicated that chemical precipitation, another chemical process, can be effectively used for the decontamination of radioactive wastewater. Its main advantages include simplicity and applicability to large volumes of wastewater. Osmanlioglu investigated the performance of potassium ferrocyanide, nickel nitrate, and ferrum nitrate as chemical agents for the precipitation of Cs-137, Cs-134, and Co-60 [67]. Laboratory jar tests revealed decontamination factors of 66.5, 8.6, and 9 for Cs-137, Cs-134, and Co-60, respectively. Only 0.98 m^3 of radioactive sludge was left after treatment from a 35 m^3 feed solution, indicating a 97.2 % volume reduction. The author also found that potassium ferrocyanide could be used in a two-staged precipitation process for large-scale applications. A recent study by Gusa et al. (2020) used barium sulfate for the decontamination of Ra-226 via co- or post-precipitation methods, and optimized the Sr/Ba molar ratio in treated water for maximum Ra^{2+} removal [68]. Similarly, insoluble sulfate minerals were explored as co-precipitating agents for the removal of Ra-226 [69]. Moreover, the combination of flocculant $Ni(NO_3)_2$ and $K_4Fe(CN)_6$ was studied for its co-precipitation effect on Cs^+ [70]. Interestingly, results showed that the precipitation process was dominated by flocculant dose rather than process conditions such as pH, temperature, and settling time. Despite not being addressed by many researchers, this might be advantageous when dealing with real nuclear wastewater where varying such conditions is faced by limitations.

Table 2
Adsorption capacities and experimental conditions of sorbent materials used for radionuclide removal.

Adsorbent	Radionuclides	Experimental conditions (C_o, t, pH)	Adsorption capacity (mg g^{-1})	Reference
BDC/MoS_2-PO_4	Uranium (VI)	C_o = 20–150 mg L^{-1}, t = 30 min, pH = 6.0	161.29	[24]
NAg_2O	Cobalt Zinc	C_o = 0.1–2.0 mol L^{-1}, t = 60 min, pH = 7.0	138.9 391.2	[35]
3-ATAR	Uranium (VI)	C_o = 2.595–26.20 μg L^{-1}, t > 24 h, pH = 7.3–8.3	98.90	[50]
U/Sr-IP2	Uranium (VI) Strontium (II)	C_o = 10–40 mg L^{-1}, t ~ 100 min, pH = 3.0–7.0	317 160	[57]
APO-10	Uranium (VI)	C_o = 3.0–35.0 mg L^{-1}, t = 60 min, pH = 5.5	826.44	[58]
KZTS	Strontium ion	C_o = 5–100 mg L^{-1}, t = 10 min, pH = 3.0–11.0	19.3	[39]
MSP	Cesium ions	C_o = 50–1200 mg L^{-1}, t = 60 min, pH = 6.0	78	[51]
KZTS-NS	Cobalt	C_o = 100 mg L^{-1}, t = 12 h, pH = 5.3	82.22	[40]
Zr-EDTMP	Uranium (VI)	C_o = 50 mg L^{-1}, t = 60 min, pH = 5.0–6.0	317.4	[59]
AO-XZ	Cesium ions	C_o = 10–100 mg L^{-1}, t = 20 min, pH = 7.0	222.2	[60]
Mesoporous graphite-C_3N_4	Thorium (IV)	C_o = 150–450 mg L^{-1}, t = 30 min, pH = 4.0	196.08	[53]
HAP aerogel	Uranium (VI)	C_o = 100 mg L^{-1}, t = 10 min, pH = 4.0	2087.6	[25]
PBA_{Cu}	Cesium	C_o = 20 mg L^{-1}, t = 30 min, pH = 5.0–9.0	97.78	[61]
C4BisC6/MMCs-P-5	Cesium	C_o = 5 mmol L^{-1}, t = 180 min, [HNO_3] = 3 mol L^{-1}	22.72	[62]
U/SBA-15-4	Uranium	C_o = 5–300 mg L^{-1}, t = 30 min, pH = 5.0	231.5	[45]
Fe_3O_4@MCM-41-PDA/OA	Uranium (VI) Europium (III)	C_o = –, t = 60 min, pH = 5.0	38.11 25.46	[46]
HTO/CFs	Uranium	C_o = 2.5 mmol L^{-1}, t = 240 min, pH = 5.0	328.24	[34]
MCM-Zr-TBP	Europium (III)	C_o = 200–1300 mg L^{-1}, t = 20 min, pH = 4.2	147.06	[47]
Fe_3O_4@Ti_3C_2-PDA/OA	Uranium (VI) Thorium (IV)	C_o = 4–50 mg L^{-1}, t ~ 20 min, pH = 5.0, 3.0	165.9 202.7	[41]
ACM	Uranium	C_o = 100 ppm, t = 24 h, pH = 7.0	390	[26]
PAN_{PA}	Uranium (VI)	C_o = 350 mg L^{-1}, t = 60 min, pH = 7.0	459.27	[49]
NiSiO@NiAlFe LDHs hollow spheres	Cesium	C_o = 5–180 mg L^{-1}, t = 20 min, pH = 10.0	61.5	[30]
PP-*g*-PAAPB	Cesium		51.38	[52]

(*continued on next page*)

Table 2 (*continued*)

Adsorbent	Radionuclides	Experimental conditions (C_o, t, pH)	Adsorption capacity (mg g^{-1})	Reference
ACF	Uranium (VI)	C_o = 10–1000 mg L^{-1}, t = 60 min, pH = – C_o = 25–200 mg L^{-1}, t = 240 min, pH = 7.0	101.2	[63]
Zr-AMTP-2	Lutetium	C_o ~ 100 mg L^{-1}, t = 6 h, pH = 6.0	113.91	[64]
CMC/IA gel	Strontium	C_o = 20–1000 mg L^{-1}, t = 2 min, pH > 4.0	144.73	[27]
$Ti_3C_2T_x$ Mxene	Cesium ions	C_o = 2 mg L^{-1}, *t* = 60 min, pH = 7.0	148	[42]
0.2PCMK-3	Uranium	C_o = –, *t* = 35 min, pH = 5.0	405.5	[65]
COF-PDAN-AO	Uranium	C_o = 258 mg L^{-1}, *t* = 25 min, pH = 1.0–4.0	410	[44]
ZrO_2@PCs-PO_4	Uranium (VI)	C_o = 50 mg L^{-1}, *t* = 30 min, pH = 5.5	496.5	[43]
PHCP-2	Uranium	C_o = 60 mg L^{-1}, *t* = 5 min, pH = 7.0	297.14	[55]
KZnFC/Al-MCM-41	Cesium	C_o = 5 mM, *t* = 10 min, pH = 2.0–11.0	154.2	[48]
HDPy-bent	Technetium-99	C_o = –, t = 3 min, pH = 2.0–10.0	57.5	[28]
CAA@MgAlFe	Uranium (VI)	C_o = 20 mg L^{-1}, *t* = 2.5 min, pH > 10.0	532.5	[29]
	Cesium (I)	C_o = 20 mg L^{-1}, *t* = 1.5 min, pH > 4.0	57.6	
EAA-AO	Uranium	C_o = 100 mg L^{-1}, t = 60 min, pH = 4.0	170.76	[56]
FP1	Uranium (VI)	C_o = 60 mg L^{-1}, *t* = 120 min, pH = 5.0	704.23	[66]

4.2. Physical processes

4.2.1. Evaporation

Evaporation, illustrated in Fig. 3, is critical in the treatment of radioactive wastewater as it significantly contributes to volume reduction. Literature pertaining to evaporation-based approaches strongly suggest its utilization for radioactive wastewater treatment. Evaporation handles the enrichment of radionuclides which is required to meet high-quality regulations. Although evaporation systems are highly effective, free of secondary contaminants, and reliable, their wide employment is challenged by specific energy requirements. Lower energy consumption evaporation technologies such as mechanical vapor recompression (MVR) and solar-driven interfacial evaporation have been researched recently.

MVR is an energy-efficient heat pump evaporation technology, where heat is recovered from the generated secondary vapor during evaporation. When the wastewater solution is evaporated, the formed vapor is compressed to enhance its energy grade. With compression, a higher temperature is attained and used to aid in solution evaporation. The operating principle of MVR allows for its low electricity consumption through a highly energy-efficient system. Additionally, MVR systems are often not labor-intensive and are compact in structure. Hou et al. (2022) developed a lab-scale MVR system to investigate its performance in the treatment of radioactive wastewater [71]. In their study, the two optimized parameters were evaporation temperature and heat transfer temperature difference. Their experiments concluded that an evaporation temperature of 90 °C produced the highest decontamination effect on strontium, cobalt, and cesium ions. Since MVR is highly energy-efficient, system efficiency was also studied by Hou et al. (2022). Their results revealed that efficiency increased as the evaporation temperature increased from 70 to 100 °C. Yu et al. (2020) explored solar-driven radioactive wastewater treatment for the removal of strontium, cobalt, and cesium [72]. The research group utilized a monolithic sponge with a three-dimensional porous structure as the evaporation platform. Under one sun irradiation, the results revealed a high evaporation rate of 1.6 kg m^{-2} h^{-1} in addition to the significant reduction in strontium, cesium, and uranium concentrations from gram to microgram levels.

4.2.2. Membrane separation

Membrane separation technology shows several advantages in the treatment of radioactive wastewater in terms of high decontamination factors, low energy consumption, and facile operation. Recently, a broad range of studies have been conducted on radioactive wastewater treatment exploring the performance of reverse osmosis (RO), forward osmosis (FO), ultrafilration (UF), membrane distillation (MD), and nanofiltration (NF) systems on radioactive wastewater treatment. Table 3 summarizes recent advances in the treatment of radioactive wastewater using membrane separation. An RO process involves stopping or reversing the flow of water molecules in a solution with a higher concentration (feed solution). In the event that the applied pressure differential is greater than the osmotic pressure difference (2–10 MPa) across a semi-permeable non-porous membrane, water molecules move from a high-solute area to a low-solute area [5]. RO membrane technology has shown great advancements in processing brackish water, seawater desalination, and wastewater. Recently, RO has been effectively applied to treat radioactive wastewater for removing radionuclides such as cobalt, cesium, and strontium. While UF has been seen in several NPPs, they are often used for the removal of macromolecules as a pretreatment step to RO units [73]. Because of RO's high rejection and low energy consumption, the removal of radionuclides using RO membranes has seen great research interest. Because the normally required decontamination factors of nuclides in radioactive wastewater reach up to 10^3–10^4, and the commercially available RO membranes cannot attain such efficiencies, research interest in modifying RO membranes has been seen. In two publications, Chen et al. (2020) studied various modified RO membranes grafted with EPTAC, PEI, PVA, and PAA [74,75]. As a result, their findings showed membrane surface charge to be a powerful parameter directly influencing the radionuclide mass transfer process. From their results, it was concluded that positively charged RO membranes are likely to obstruct the radionuclide mass transfer by electrostatic repulsion, as demonstrated in Fig. 4. Thus, appropriate modification of RO membranes along with suitable characterizations with regards to zeta potential analysis is key to the successful implementation of RO membranes for radioactive wastewater. In addition to surface charge, fouling resistance and selectivity of RO modified membranes are critical parameters to investigate as well. In that regard, the same research group studied the effect of surface properties on surfactant fouling which deteriorated RO membrane flux by 35–75 % [76]. Interestingly, hydrophilic neutral charged membranes demonstrated the lowest surfactant resistance, providing the greatest selectivity.

Furthermore, Li et al. (2018) designed a disk tubular reverse osmosis (DTRO) system for the treatment of radioactive wastewater generated in a high-temperature gas-cooled reactor (HTGR). Such reactors, designed by the Institute of Nuclear and New Energy Technology, are considered advanced nuclear processors. Thus, the study explored the feasibility of radioactive wastewater treatment from a real application in pilot scale.

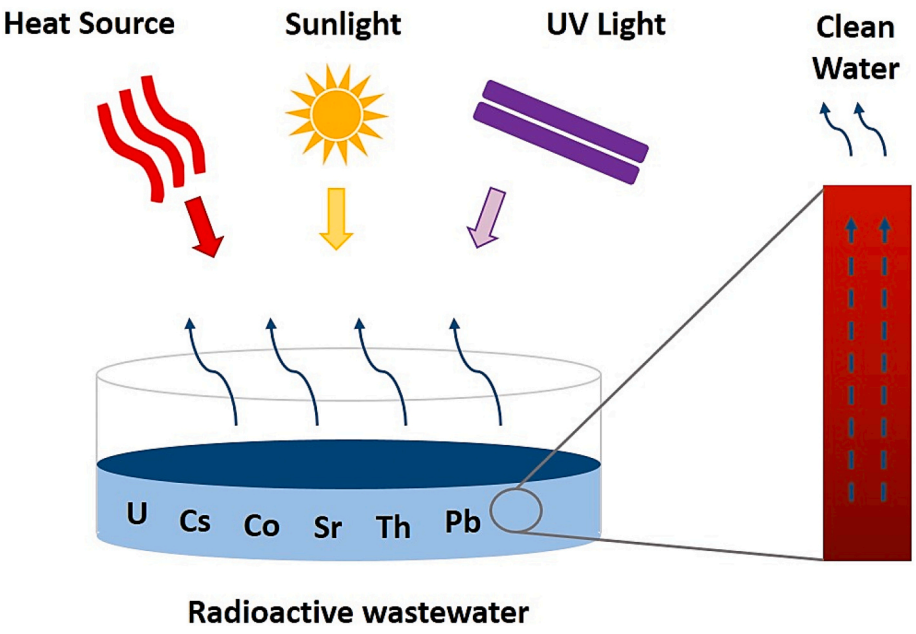

Fig. 3. Schematic illustration of evaporation for the treatment of radioactive wastewater.

With a volume reduction factor of 50 and a decontamination factor of 5760, the authors confirm the suitability of the designed DTRO system for the treatment of low and intermediate radioactive wastewater, as well as wastewater from HTGR [77]. While extensive research on RO membranes have demonstrated radionuclide rejection of up to 98 %, NF processes are less energy intensive with a higher permeate flux. A study by Chen et al. (2018) investigates the performance of different NF membranes in the removal of strontium-containing radioactive wastewater [78]. Commercial NF 270, NF 90, and XN 45 membranes were utilized to study the effects of initial strontium concentration, solution pH, and complexation phenomena. The findings suggest enhanced rejection as initial strontium concentration decreased, favoring their application in low-contaminated radioactive wastewaters. Moreover, minimum rejection was obtained at the membrane's isoelectric point of pH 5. Finally, polyacrylic acid and ethylenediamine tetraacetic acid disodium salt were introduced as complexing agents which were found to improve rejection rates of the membranes. In their study, the research group also attained rejection rates as those of the virgin membranes after cleaning with DI water and citric acid.

There have been numerous studies to develop forward osmosis (FO) processes for radioactive wastewater treatment, as they exhibit the advantages of high retention, water flux, and low energy consumption. Unlike pressure-driven membranes, FO processes eliminate the need for an external energy source due to the absence of high-pressure devices by transferring water molecules from a low-osmotic feed solution to a high-osmotic draw solution; FO uses the difference in osmotic pressure [5]. With FO technology, the ever-lasting bottleneck in draw solution optimization faces its wide application. However, when it comes to radioactive wastewater treatment, this problem is less apparent. Most NPPs are located near coastal cities, where seawater is easily accessible. As such, the draw solution is easily obtained, as well as effortlessly regenerated in case of high radionuclide retention. Diluted seawater is directly discharged into the sea post FO treatment. When using FO membranes, performance is highly influenced by membrane properties, feed solution quality, draw solution quality, and hydrodynamic conditions. Additionally, FO can be operated in two modes, with active layer facing feed solution (AL-FS) or active layer facing draw solution (AL-DS). Depending on the radionuclide targeted for removal, the mode is chosen. Radionuclide solute diffusive flux occurs by convection or due to transmembrane nuclide concentration.

In a research paper by Liu et al. (2018), the authors studied CTA-NW, CTA-ES, and TFC-ES as FO membranes, while varying operating conditions [79]. The group reported the highest cesium rejection when working with the CTA membrane and in AL-FS mode. Flow velocity was found to not affect retention in either operating modes. The authors further explored the effect of commercial FO membranes on multi-nuclides in a more recent study [80]. It was apparent that the diffusion coefficient of nuclides and reverse salt flux left a much higher impact on nuclide flux, unlike water flux, salt concentration, flow velocity and partition coefficient. Furthermore, borate-containing radioactive wastewater is produced in NPPs, which signifies research efforts towards the removal of radionuclides from borate-containing radioactive wastewater [81]. FO was used for the decontamination of borate-containing radioactive wastewater. Membrane property played a key role where the embedding of polyester screen support in cellulose triacetate (CTA-ES) established the highest cobalt and strontium ions retention with borate retention <37 %. Modified RO membranes have also been used to separate silicon and radionuclides from boron-containing radioactive wastewater. When using PEI grafting to modify commercial membranes, high radionuclide rejection and low boron rejection were obtained [82].

The long-operation of FO processes in the treatment of radioactive wastewater has been found to face an important problem which is fouling, due to the deposition of radionuclides such as Co(II), Sr(II), Cs (I), and Na(I) on both the active and support layer of the FO membranes. As a result, the membrane surface characteristics change, exhibiting a surface higher in roughness and hydrophobicity. The on-line cleaning of FO membranes with DI water and external cleaning by ultrasound and HCl were applied by Liu et al. (2020) [83]. Results revealed that a water flux recovery of 69 % was possible with the reported cleaning approach.

Membrane distillation (MD) is an emerging membrane technology in which the hydrophobic nature of the membranes only allows the passage of volatile substances such as water vapor, leaving behind non-volatile substances to be concentrated in the feed side. In MD, processing requires mild operating conditions and a low emphasis on chemical interactions between membrane and feed solutions. MD produces pure water that can be reused or discharged into the environment safely. Unlike normal evaporation, MD avoids corrosion, scaling, or foaming, as well as droplet entrainment. By achieving the high concentration of solute in a single step and avoiding the sorption of ions like Co^{2+}-50,

Table 3
Membrane technologies and experimental conditions used for radionuclide removal.

Technology/membrane material	Radionuclide	Feed/process conditions	Performance	Reference
VMD/PTFE	Uranium	Real low-level radioactive wastewater (LLRW) Temperature = 75 °C Feed flow velocity = 0.7 m s^{-1} Vacuum pressure = 0.01 MPa pH = 5	Permeate flux = 11.3 LMH Rejection >99.99 % Concentration factor = 9	[86]
VMD/PTFE-CPVC	Cesium ions (Cs^{+})	Simulated wastewater Temperature = 70–90 °C Vacuum pressure = 0.01–0.03 MPa	Permeate flux = 1.59–6.82 kg m^{-2} h^{-1} Max decontamination factor = $10^{4.85}$ Concentration factor = 110.3	[87]
VMD/PP	Cobalt ions (Co^{2+})	Simulated wastewater Inlet concentration = 10 mg L^{-1} Temperature = 30–70 °C	Rejection = 99.67 % Permeate flux = 6.3 LMH	[88]
NCMD/PTFE	Strontium ions (Sr^{2+})	Simulated wastewater Total α and β activity concentrations = 21,279.7 and 17,811.9 Bq L^{-1} Inlet air temperature = 44.9–77.7 °C	Rejection = 99.99 % Decontamination factor = 4.5 × 10^{6} Permeate flux = 5.21–24.87 kg m^{-2} h^{-1}	[89]
DCMD/PP	Strontium (II) Cobalt (II) Cesium (I)	Simulated wastewater Feed temperature = 40–80 °C	Permeate flux = 2.7–29.2 LMH Decontamination factor > 10^{5}	[84]
DCMD & AGMD/PVDF	Cesium ions (Cs^{+}) Cobalt ions (Co^{2+}) Chromium (Cr^{3+})	Simulated wastewater Inlet concentration = 450 ppm of each nuclide Transmembrane pressure = 1 bar Feed flowrate = 0.75 L min^{-1} Feed temperature = 70 °C	**DCMD** Permeate flowrate = 11.24 kg m^{-2} h^{-1} *DF*: 22 × 10^{2} 96 × 10^{3} 62 × 10^{3} **AGMD** Permeate flowrate = 7.51 kg m^{-2} h^{-1} 15 × 10^{2} 69 × 10^{3} 26 × 10^{3}	[85]
NF/PA	Strontium	Simulated wastewater pH = 4–9 Inlet strontium concentration = 5–30 mg L^{-1} Operating pressure = 0.5 MPa	Rejection = up to 100 % Permeate flux = 85.7 LMH	[78]
RO/PA-EPTAC	Cesium (I) Cobalt (II)	Simulated wastewater Inlet concentration = 2000 μg L^{-1} of each cesium (I) and cobalt (II) Transmembrane pressure = 1.20 MPa	DF: 151.12 1670.84	[74]
RO/PA-(EPTAC, PEI, PVA, PAA)	Cesium ions (Cs^{+})	Simulated wastewater Inlet concentration = 2000 μg L^{-1} Transmembrane pressure = 1.20 MPa	Flux = 51.42–96.10 LMH	[75]
RO/PA-(EPTAC, PEI)	Cesium (I) Cobalt (II)	Simulated wastewater Inlet concentration = 2000 μg L^{-1} Cs (I), Co(II). 500 μg L^{-1} B.	Flux = 51.42–53.58 LMH	[76]
RO/PA-PEI	Cesium (I) Cobalt (II)	Simulated wastewater Inlet concentration = 2000 μg L^{-1} Cs (I), Co(II). 500 μg L^{-1} B. 8 mg L^{-1} Si.	DF: 1230.78 96,352.10	[82]
DTRO	HTGR radioactive wastewater	Real HTGR radioactive wastewater Pressure = 3 Stage (2.2, 1.8, 3.1 MPa)	Volume reduction factor = 50 DF = 5760	[77]
FO/CTA-ES	Cobalt (II) Strontium (II) Cesium (I)	Simulated wastewater Inlet concentration: 20 mg L^{-1} $CoCl_2$, 20 mg L^{-1} $SrCl_2$ and 20 mg L^{-1} CsCl Recirculation = counter-current Flow velocity = 2–11 cm s^{-1} Orientations = AL-FS and AL-DS Draw solution = 0.5–2 M NaCl	Nuclide flux (AL-FS): 1.54 mg m^{-2} h^{-1} 10.22 mg m^{-2} h^{-1} 15.63 mg m^{-2} h^{-1} AL-FS nuclide flux < AL-DS nuclide flux	[80]
FO/CTA-NW, CTA-ES, PA-ES	Cesium (I)	Simulated wastewater Inlet concentration: 0.15 mmol L^{-1} (20 mg L^{-1}) Cs(I). Draw solution = 1.5 M NaCl Flow velocity = 11 cm s^{-1}	**CTA-ES:** Cs (I) retention = 96.24 % Water flux = 33.34 LMH AL-FS nuclide flux < AL-DS nuclide flux	[79]
FO/CTA-NW, CTA-ES, PA-ES	Cobalt (II) Strontium (II) Cesium (I)	Simulated wastewater Inlet concentration: 20 mg L^{-1} of $CoCl_2$, $SrCl_2$, CsCl Boric acid: 0, 800, 1600–2400 mg L^{-1} Draw solution = 1 M NaCl	Retention (CTA-ES): Co^{2+} = 99.62–99.69 % Sr^{2+} = 96.90–97.85 % Cs^{+} = 95 % AL-FS retention > AL-DS retention	[81]
UF/PVDF-PVP-$K_2S_2O_8$	Cobalt (II) Strontium (II) Cesium (I) Silver (I)	Simulated wastewater Inlet concentrations: 0.1 mg L^{-1} of Cs(I), Co(II), Sr(II) and Ag(I) and 200 mg L^{-1} concentration of SDBS	Flux = 170.7 LMH Rejection of Sr(II) and Co(II) >98 %	[73]

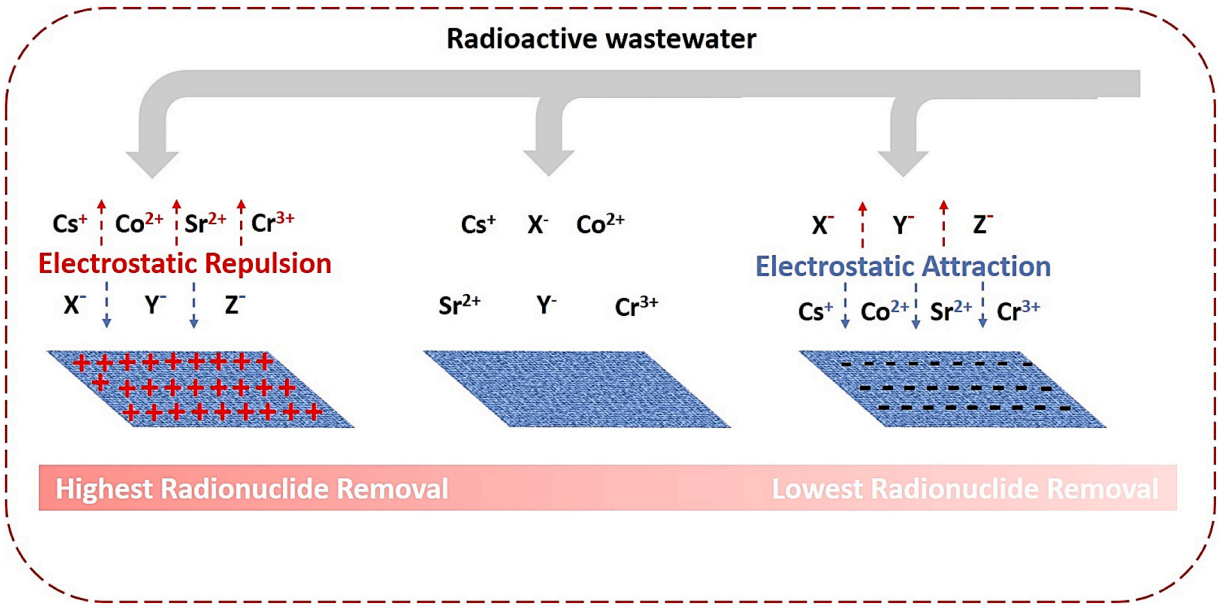

Fig. 4. Membrane rejection of radionuclides based on electrostatic repulsion.

Cs^+-137, and Cs^+-134 inside the membrane pores, MD is able to achieve the high solute concentration necessary for fossilization in concrete up to 25 % of solute [4]. Operation at a low evaporation temperature can decrease the volatility of some volatile nuclides present in the waste, such as tritium, some forms of iodine, and ruthenium. MD systems can be operated in multiple configurations such as direct contact membrane distillation (DCMD) [84], air gap membrane distillation (AGMD) [85], vacuum membrane distillation (VMD) [86–88], and sweeping gas membrane distillation (SGMD). While these are conventional MD configurations, a novel one, non-contact MD has been developed by Li et al. (2022) [89]. An in-house developed non-contact MD method was proposed to mitigate the drawbacks of fouling, wetting, short half-life, and low flux of conventional MD configurations [89]. VMD has been the most utilized configuration for the decontamination of radioactive wastewater. Jia et al. (2018) utilized VMD for the removal of cobalt ions from simulated radioactive wastewater [88]. In their study, the research group varied operational parameters to explore their effects on the mass transfer mechanism of commercial polypropylene hollow fiber membranes. Findings propose permeate vacuum, among other parameters such as feed temperature and feed flow rate, to be the most impactful on membrane flux. With an initial Co(II) concentration of 10 mg L^{-1} and a permeate side vacuum degree of 0.9 atm, the VMD process exhibited a removal efficiency of over 99.67 % towards Co(II) ions. The mass transfer process of multiple radioactive wastewater pollutants in VMD systems was also studied by Dragoi and Vasseghian (2020). Feed temperature, permeate temperature, and permeate pressure were studied by the means of artificial neural networks (ANNs).

Although VMD has been studied by a few researchers for the treatment of radioactive wastewater, they have mostly prepared artificial feed solutions in their studies. As a result, the complex composition and actual radioactivity nature of real radioactive wastewater solutions remain unexplored. Nie et al. (2021) were among the first to study the feasibility of VMD systems for the treatment of uranium contained LLW from UO_2 fuel element industry [86]. In their study, commercial tetrafluoroethylene (PTFE) hollow fiber membranes were used to investigate operational parameters such as feed temperature, pH, vacuum pressure, and feed flow velocity. Using real feed solutions, the researchers reported the maximum feasibility of VMD with the determined optimum operational parameters of a feed temperature of 75 °C, feed flow velocity of 0.7 m s^{-1}, a vacuum pressure of 0.01 MPa, and pH of 5. Under those conditions, the VMD system achieved a permeate flux of 11.3 LMH and a rejection >99.99 %. Moreover, Jia et al. (2021) explored a pilot-scale VMD system to ensure the reproducibility of bench-scale experiments on an industrial scale [87]. The study showed VMD to be a highly promising candidate for the wide application in radioactive wastewater treatment. Despite the successful exploration of real radioactive wastewater and pilot-scale systems in VMD processes in two consecutive studies, the investigation of real radioactive wastewater in pilot-scale VMD systems has not been discovered yet.

4.3. Electrochemical processes

Recently, electrically driven separation processes, such as electrosorption, electrodialysis, and electrodeionization, have been used for the treatment of radioactive wastewater. A schematic illustration of electrodialysis and electrosorption for the treatment of radioactive wastewater is shown in Fig. 5. Electrochemical technologies are known for their environmental compatibility and mild operating conditions [90]. Due to the high energy efficiency, facile operation, and simple regeneration of electrosorption, it has been favorably researched. Since the performance of electrosorption is highly dependent on the electrode materials, various research studies have investigated electrode materials such as activated carbon, carbon/polypyrrole, floriform WO_3/C, and zinc. Yu et al. (2022) prepared biomass-derived carbon/polypyrrole electrodes for the electrosorption of U(VI) [91]. Biomass-driven carbon was also functionalized and used as an electrode for the removal of another radionuclide, strontium. A hybrid membrane capacitive deionization (CDI) system was coupled with a porous activated carbon (PAC) anode and an anion-exchange membrane. As such, the PAC provided good electrical conductivity and sufficient surface area whereas the anion-exchange membrane provided selectivity towards strontium [92].

Flow electrode capacitive deionization (FCDI) is often used to concentrate radioactive wastewater. This solution addresses problems with insufficient storage spaces in nuclear plants. Zhou et al. (2022) ran batch experiments to investigate the performance of FCDI on the removal of uranium from radioactive wastewater [93]. In their study, the research group reported the accumulation of uranium into the electrolyte and on the flowing activated carbon-containing electrode thus presenting FCDI as a promising approach to radioactive wastewater treatment. Previously, the same research group addressed pseudocapacitive deionization with a WO_3/C electrode [94]. In that study, conventional CDI was investigated, where limitations with regard to the

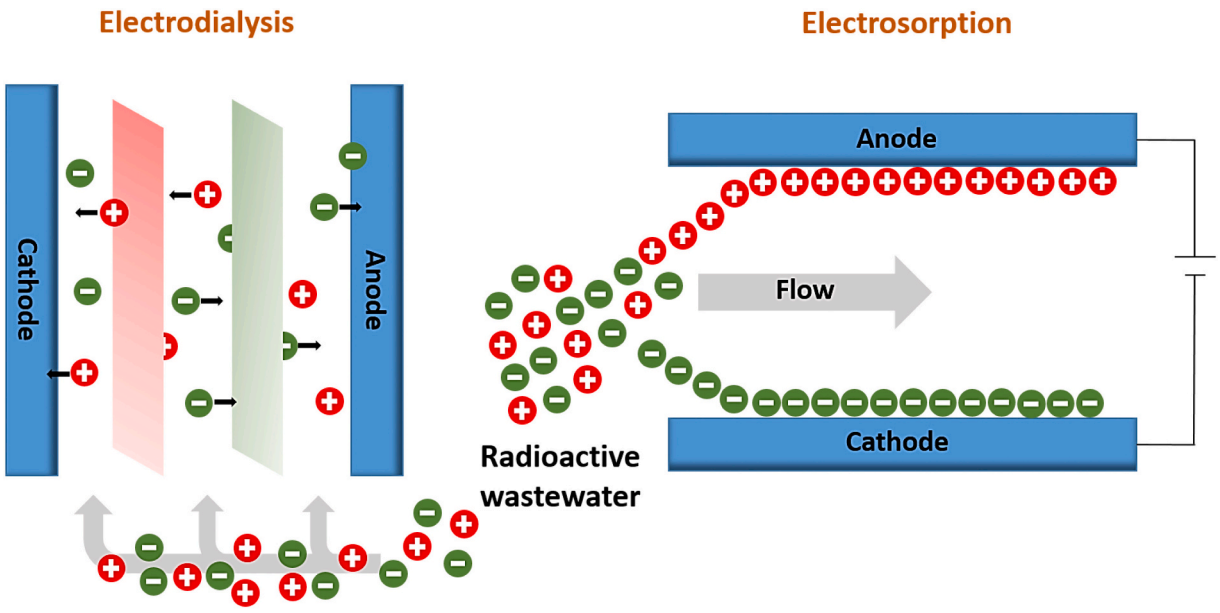

Fig. 5. Schematic illustration of electrodialysis and electrosorption for the treatment of radioactive wastewater.

amount of immobilized active material and intermittent adsorption/desorption processes. Therefore, FCDI emerges as a promising alternative, boosting production efficiency and scale-up feasibility.

Alternatively, Yang et al. (2021) use the in situ electrochemical synthesis method (ISESM) of CsZnFC to treat LRAW [95]. While ISESM of CsZnFC has been explored for other applications, this study reports its first utilization to remove cesium from radioactive waste. Notably, the authors report a removal rate of up to 99.42 %, leaving behind a residual concentration of 29.13 μg/L. Moreover, the electrochemical mineralization method was utilized by Lv et al. (2021) for the rapid purification of uranium-containing wastewater (UCW) [96]. While magnetite was crystallized and generated in UCW, uranium was simultaneously incorporated. In their study, metallic iron was the only required raw material with no secondary pollutant formed. Not only did the method remove uranium from UCW effectively, but the isolation of uranium, in the form of uranium oxide, was also successfully attained.

4.4. Biological processes

The decontamination of radioactive wastewater using biological processes has always been considered a viable alternative to chemical processes and adsorption mechanisms as it presents a clean, low-cost, effective, and easy processing approach to the removal of radioactive contaminants. This has been discussed by a great number of authors in literature. In fact, literature reviews have discussed a number of methods to treat radioactive contamination in soil and surface water [1]. These include microbial bioremediation and plant bioremediation techniques such as phytoextraction, phytovolatilization, phytostabilization, rhizofiltration, and phytodegradation. In applications targeting the biological treatment of radioactive wastewater, however, this review shows that biosorption and bioaccumulation are the most widely investigated. While the uptake of microorganisms by biosorption is metabolism-independent, bioaccumulation is characterized by its metabolism dependence (i.e., the need for cellular energy). Table 4 summarizes recent advances in the treatment of radioactive wastewater using biosorption and bioaccumulation.

4.4.1. Biosorption

Amid various conventional treatment approaches used to treat radioactive wastewater, adsorption has been the most researched and

Table 4
Biomedia used and their biosorption and bioaccumulation effects on for radionuclide removal.

	Biomedia	Target radionuclide	Kinetic model	Sorption isotherm	Biosorption capacity (mg g^{-1})	Reference
Biosorption	Alginate-immobilized *Aspergillus niger*	Thorium ions	Pseudo-second-order	Freundlich	303.95	[98]
	Magnetic cyanoethyl chitosan beads	Cobalt (II)	Second-order	Langmuir	17.92	[101]
	Ion-imprinted honeycomb-like chitosan/kaolin clay composite foams	Uranium (VI)	Pseudo-second-order	Langmuir	286.85	[97]
	Active and inactive *Aspergillus niger*	Uranium (VI)	Pseudo-second-order	Langmuir	Active 50.65 Inactive 83.40	[99]
	Trichoderma harzianum mutant	Uranium (VI)	Pseudo-second-order	Langmuir	83.59	[100]
	Microcystis aeruginosa	Uranium (VI)	Pseudo-second-order	Langmuir	51.79	[102]
Bioaccumulation			Bioaccumulation efficiency (%)		Bioaccumulation factor	
	Haematococcus strains	Cesium	>50		–	[103]
	Ludwigia stolonifera	Cobalt	>95		37.2	[104]
		Cesium	>65		42.4	
	Desmodesmus armatus	Cesium	>95		–	[105]
	Myriophyllum spicatum	Cobalt	>90		10.80	[106]
		Cesium	>60		27.13	

thus is considered highly promising. However, due to the high cost of manufactured adsorbents, their wide application is challenged. Biosorption emerges as an eco-friendly and inexpensive substitute which stands out at resource utilization. The selective biosorption of U(VI) from aqueous solution was explored by Yu et al. (2022) [97]. The research group developed ion-imprinted honeycomb-like chitosan/kaolin clay (ICK) composite foams as the biosorbent. The results revealed a sorption capacity up to 286.85 mg g^{-1} at 298 K and pH 5, with the collected kinetic data following a pseudo-second-order model. In this study, both amine and hydroxyl groups enhanced the U(VI) removal from the aqueous solution.

Ding et al. (2019) prepared a novel alginate-immobilized *Aspergillus niger* microsphere (AAM) biosorbent for the removal of Thorium (IV) [98]. Rapid complexation of the biosorbent with Th (IV) was observed at pH 6.0 and 40 °C. Increased surface area and biosorption sites contributed to biosorption capacities as high as 303.95 mg g^{-1}. In addition to its advantages as a low-cost and eco-friendly biosorbent, the developed material exhibited good recyclability as well. Another research group also examined the effect of *Aspergillus niger* on the treatment of radioactive wastewater [99]. In their study, the researchers explored the efficiencies of both active and inactive *Aspergillus niger* on the biosorption capacity of U(VI). Interestingly, the maximum biosorption capacities found for active and inactive *Aspergillus niger* were 50.65 mg g^{-1} and 83.40 mg g^{-1} at pH 5. While inactive microbes often show better biosorption capacities than active ones, their overall strength is significantly reduced and thus, the structure loosens, reducing utilization and recycling rates. Similarly, researchers have explored several mutation strategies to enhance the biosorption performance of microorganisms. Liang et al. (2021) applied a cold atmospheric plasma jet (CAPJ) to mutate *Trichoderma harzianum*, which resulted in a higher biosorption capacity of U(VI) than raw *Trichoderma harzianum* [100].

It was observed that most of the reviewed biosorption studies with regards to the removal of radionuclides followed the Langmuir isotherm and pseudo-second-order model, indicating that that the biosorption process was primarily controlled by chemical adsorption or ion exchange. Moreover, the proper functionalization of biomass played a vital role in the sorption capacity towards the targeted radionuclide.

4.4.2. Bioaccumulation

The bioaccumulation of radionuclides in microorganisms is described by their transport from the cell surface into the cell. The bioaccumulation factor (BCF) is used to describe the radionuclide concentration in plant tissue divided by initial concentration in water.

$$\mathrm{BCF} = \frac{\text{Activity content in dry mass weight, Bq/g}}{\text{Activity content in spiked water, Bq/ml}}$$

A local aquatic plant in Egypt, Ludwigia stolonifera, was recently explored for its bioaccumulation efficiency in the removal of ^{60}Co and ^{137}Cs. It was observed that the plant could accumulate up to 95 % and 65 % of ^{60}Co and ^{137}Cs, respectively, in a simulated wastewater feed solution. Kim et al. (2021) also investigated the performance of two newly identified Haematococcus strains for the bioaccumulation of radioactive cesium. The discovered haematococcus strains were successful in the removal of up to 50 % of radioactive cesium from low-level contaminated water containing 5 Bq/ ml initial concentration.

Both ^{60}Co and ^{137}Cs are gamma emitters which are commonly discharged under normal operating conditions in nuclear installations. Saleh et al. (2020) investigated an innovative approach to the decontamination of radioactive cobalt and cesium by utilizing submerged plant, *Myriophyllum spicatum*, with no agricultural benefit. In fact, *Myriophyllum spicatum* is considered a threat for the aquatic environment. In their study, Saleh et al. (2020) optimized various parameters such as contact time, pH, illumination, and biomass to achieve 90 % and 60 % removal rates of cobalt and cesium, respectively. ^{137}Cs, in particular, have caused major concern following the Fukushima Daiichi NPP accident in 2011. Thus, Kim et al. (2019) studied the bioaccumulation effects of nine different microalgae on radioactive cesium. Among the microalgae investigated, they report *Desmodesmus armatus* to exhibit the highest bioaccumulation efficiency. This study combines bioaccumulation and magnetic separation to achieve >90 % removal of the radioactive cesium from an aqueous solution.

In general, current research on the bioaccumulation of radioactive metals via local, submerged, or newly discovered plants adds great value to natural resource utilization. Additionally, they present natural phytoremediation tools after nuclear accidents or uncontrolled applications of artificial radioisotopes.

5. Evaluation of conventional treatment technologies

It is realized from the above literature review on conventional treatment technologies used for radioactive wastewater and from Fig. 2 (b) that the majority of research groups focused on adsorption and membrane separation. A great decontamination potential has been seen with adsorption, with various different types of adsorbents explored. Although naturally-derived adsorbents are usually highly abundant and economical, they are found to possess low adsorption capacities. Thus, natural adsorbents have only been assessed to a limited extent, and most researchers have either (i) explored the modification of naturally abundant adsorbents, or (ii) developed engineered adsorbents for the removal of radionuclides. Traditional adsorbents such as clays and LDHs possess slow kinetics and limited selectivity. However, recent modifications have demonstrated improved kinetics leading to low requirements in contact time. Advances in carbonaceous adsorbents had good adsorption capacities, however, the incorporation of carbon materials in MOFs and COFs outperformed other structures due to their highly porous three-dimensional structures. Engineered adsorbents are generally difficult to prepare in large amounts due to their fabrication techniques. Additionally, they are not economical for scaled-up applications. Moreover, although most papers on adsorption study the effect of pH on decontamination performance, most adsorbents show ideal behavior in conditions that do not realistically represent actual nuclear wastewater. Temperature had not been considered as much since most bench-scale experiments take place at room temperature. Although research on radionuclide adsorption has been illuminated, no study, to date, has examined the effect of radiation on adsorbents. As an ideal adsorbent needs to exhibit high removal rates, high stability in conditions that mimic those found in real applications, and demonstrate regeneration and reusability, further studies are required, particularly to design adsorbents that are stable under harsh conditions and are unaffected by radiation. Although physical/chemical modifications and fabrication of organic-inorganic hybrids have been considered as the main two pathways to improve adsorption performance [3], this review demonstrates the current research emphasis on the former.

Membrane separation shows great potential in radioactive wastewater decontamination. It is believed that ultrafiltration (UF) membranes cannot satisfactorily reject radionuclides in wastewater. Indeed, based on the review done, UF has only been suggested as a pretreatment step for other membrane units. Thus, this approach remains briefly addressed in the literature. Although NF membranes can reject multivalent ions, they have not been sufficiently studied. Therefore, previous studies on radionuclide rejection using NF cannot be considered conclusive. On the other hand, reverse osmosis (RO) has been studied by more researchers possibly due to their efficient rejection of all kinds of ions. At the same time, problems with fouling are more apparent in RO. Moreover, efforts have been perceived with emerging FO membranes for the decontamination of radioactive wastewater, with high rejection rates achieved due to the absence of hydraulic pressure. However, low flux remains a problem. Among all membrane technologies, MD has been studied the most. In radioactive wastewater treatment, MD takes advantage of the presence of low-grade heat in NPPs. This has attracted the interest of many researchers. MD does not require high operating pressures and significant chemical interactions between membrane and

feed solution. Additionally, in the presence of low-grade waste heat, MD is also considered a low-energy-consuming technology. Thus, different types of MD configurations have been studied to present MD as the most promising membrane technology for radioactive wastewater decontamination. It is important to note that in membrane technologies, radiation's effect on polymeric membranes is defeating. Thus, it is suggested to explore other types of membranes such as ceramic membranes in the future.

In studies utilizing chemical precipitation, the main challenge is within the formation of insoluble precipitates. Aside from this, the method is very simple (i.e., increasing its applicability in several wastewater types), exhibits high efficiency, and has a low cost. More importantly, few researchers have focused on process conditions such as pH, temperature, and settling time, suggesting their low impact as compared to the dosage of the utilized chemical agent. Moreover, studies using evaporation to treat radioactive wastewater showed outstanding performance in terms of volume reduction and decontamination factors. However, the method suffers from corrosion and high energy consumption. Thus, it is recommended for use on small volumes of radioactive wastewater. Several electrochemical methods have been studied as well, from which electrosorption seems to be the most promising due to its high energy efficiency, facile operation, and simple regeneration. However, when opting for electrical techniques, the presence of ions is obligatory; thus, only wastewaters with high ionic concentrations are suitable. Finally, this article reviews biological methods for radioactive wastewater treatment. Existing research has many problems in representing the drawbacks of biological methods in radionuclide removal. For instance, similar to the issues encountered with adsorbents and membranes, the impact of radiation on microorganisms is yet to be investigated and optimized. Besides, biological methods offer environmentally friendly and economic methodologies for radioactive wastewater treatment.

6. Hybrid technologies

As demonstrated in this review, numerous technologies have been recently investigated for the treatment of nuclear wastewater. These single technologies have shown great potential in wastewater treatment; however, integrating multiple technologies is expected to achieve the highest decontamination rates. Thus, several hybrid technologies have been proposed and explored by researchers as well. The most researched hybrid technology integrates membrane separation with adsorption, as depicted in Fig. 6. Bacterial cellulose membranes were modified with EDTA via a crosslinker to selectively remove Sr^{2+} from radioactive wastewater through adsorption. Strontium ions were adsorbed onto the prepared membranes due to the complexation of tertiary amines and carboxylate. Among different organic bio-adsorbents used for the removal of strontium, the adsorption capacity of the synthesized adsorbent was at the highest level [107]. Highly abundant amyloid fibers were similarly utilized for the cost-effective removal of radionuclides from water [108]. Furtherly, Kim et al. (2021) prepared an electrospun nanofiber matrix membrane with KCuHCF nano adsorbent through an in-situ immobilization step. In their work, the authors confirmed that the formation of a porous composite of selective adsorbents can serve as an effective membrane for the selective removal of radionuclides [109]. FO membranes have also been utilized for the adsorption of radionuclides from nuclear wastewater. Due to the location of most NPPs, highly abundant seawater could be used as a draw solution, after which it could be simply dumped into the sea without the need for further processing [110]. Thus, combining the power of adsorptive membranes with such FO benefits in the nuclear industry serves as a promising hybrid technology for radioactive wastewater decontamination. Likewise, other adsorptive membranes were synthesized for the selective decontamination of nuclear wastewater [111–116].

Other research groups have focused on processes integrating adsorbents with electrochemical separation techniques [91–93,117]. For

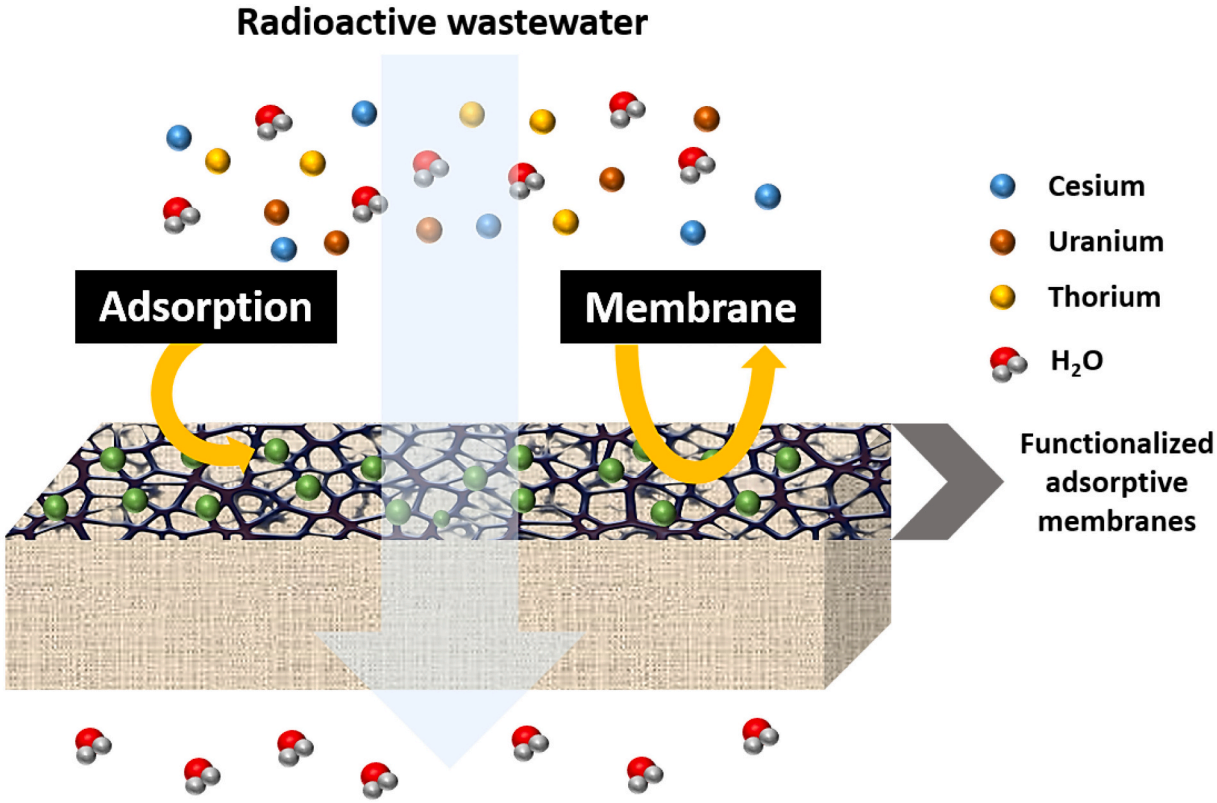

Fig. 6. Schematic illustration of the dual working principle of adsorptive membranes for the treatment of radioactive wastewater.

instance, Zhou et al. (2022) explore FCDI for the electrosorption of UO_2^{2+} [93], whereas Yu et al. (2022) use electrodeposited polypyrrole/biomass-derived carbon composite electrodes for uranium adsorption [91]. An electrochemical adsorption and desorption reaction was designed for the sequential removal of cesium using carbon nanofiber-Prussian blue (CNF-PB) composites. The highly organized structure composite exhibited strong adsorption and desorption of cesium by simply varying voltages, through a series of cycling experiments [118]. A bench-scale countercurrent two-stage adsorption-flocculation-microfiltration process was designed by Zhang et al. (2021) for the removal of divalent cobalt ions from wastewater. In the same study, the research group confirms that the hybrid system significantly enhanced the adsorption of cobalt ions with a decontamination factor of up to 1176. Not only was adsorption capacity high, but removal kinetics were also impressive, with an ability to reach equilibrium in 40 min [40]. Precipitation and filtration systems were also integrated for the removal of radionuclides. Chugunov and Vinnitskii (2020) studied dialysis membranes for the separation of radionuclides present in NPP wastewater via precipitation. Additionally, an integrated RO and MD system is suggested to be a promising hybrid technology due to the high nuclide rejection and volume reduction of radioactive wastewater [119]. All in all, by combining several single treatment processes, hybrid technologies provide combined benefits at optimized conditions. Thus, it is reasonable to conclude that hybrid technologies should remain the focal point of research interest for the continuous advancement in nuclear wastewater decontamination.

7. Reuse applications

Conventional and hybrid treatment technologies such as ion exchange/sorption, precipitation, evaporation, membrane separation, electrochemical processes, biosorption, and bioaccumulation strive at the decontamination of nuclear wastewater. As such, these treatment technologies have been significantly enhanced to comply with the required water quality standards before environmental discharge. In many industries, the recognition of wastewater as a valuable source of different compounds is increasing. With that said, research focus is shifting towards the integration of resource recovery in wastewater treatment processes. In the nuclear industry, there are concerns as to the availability of sufficient wastewater management policies to deal with the generated radioactive wastewater, making the central focus on developing appropriate management protocols in practice [120]. Thus, resource recovery is overlooked in the nuclear industry. The nuclear industry generates radioactive waste containing numerous valuable radionuclides which could have potential reuse applications. However, current efforts solely focus on safely discharging radioactive wastewater after treatment rather than extracting and recovering radionuclides. For example, MD processes have been expanded to target the discharge of treated radioactive wastewater into the environment [121]. Arnal et al. (2017) utilized an experimental pool type reactor for the decontamination of radioactive water. Their study used a two-stage RO unit followed by a low-pressure evaporator to concentrate the radioactive waste. The process achieved a high volume reduction of 600, and the produced permeate met the legal discharge limit [122].

Radionuclides could be used for their radiation and chemical properties. Among their general applications are mining, biological tracing, nuclear medicine, and equipment. For instance, nuclear medicine uses pharmaceutical products containing radionuclides to diagnose and treat disease. The most used radionuclide in such applications is Technetium-99m, Tc(99-m), a fission product of Uranium-235, and it comprises 80 % of all nuclear medicines techniques worldwide [123]. As can be seen in Table 1, Tc(99-m) is a released radionuclide from the nuclear industry. This opens up an opportunity for the recovery and reuse of Tc(99-m) in the diagnosis and treatment of diseases. This review shows that Tc(99-m) has only been targeted through adsorption using HDPy-bent, yielding an adsorption capacity of 57.5 mg g^{-1}. It is therefore worth investigating desorption capabilities of promisisng adsorbents such as HDPy-bent where radionuclides such as Tc(99-m) can be extracted and reused. Other radionuclides are also used in nuclear medicine, where cesium-137 is used in treating cancer to measure correct patient dosages of radiopharmaceuticals, cobalt-60 is used to treat cancer and sterilize surgical instruments, and Iodine-131 is used to diagnose and treat thyroid disorders including cancer and for basic biomedical research [124]. Ce(137), Co(60), and I(131) are all found in released radioactive waste, and their extraction could make their applications more cost-effective, given their source. Although no recent efforts have been noted for the removal of I(131), numerous adsorption and membrane separation solutions have been listed in Tables 2 and 3 for the removal of Ce(137) and Co(60). A desirable treatment option for patients with many micrometastases is targeted alpha therapy (TAT). It has several benefits, including as simple administration, the capacity to treat several lesions at once, and the potential to combine it with other therapy modalities for increased efficacy. TAT uses radionuclides that emit alpha particles to treat a variety of cancers, including thyroid cancer, bone tumors and metastases, hepatic metastases, ovarian cancer, neuroendocrine tumors, leukemia, lymphoma, and metastatic prostate cancer. Numerous different malignancies treated by TAT include polycythemia, cystic craniopharyngiomas, hyperthyroidism, synovitis, and arthritis. In addition, it is occasionally utilized to treat HIV, fungal, and bacterial infections. Due to Pb-212's high-linear energy transfer (LET) traversal of the nucleus and effects on cell cycle disruption and DNA damage repair, which have a significant impact on catastrophic dsDNA destruction and significantly kill tumor cells, it is the ideal alpha-emitting radiometal in terms of availability, chelation chemistry, and half-life. Pb-212 would creatively address the elusive objective of developing magic-bullet cancer treatment [125,126]. Interestingly, those radionuclides are found in nuclear waste [13,127,128], making their recovery and reuse highly beneficial and cost-effective, let alone being an approach to waste management. The removal of lead as a heavy metal using membrane separation technologies has been thoroughly investigated and validated previously [129–131]. However, no efforts have been made to validate these technologies for nuclear wastewater applications where feed compositions and process conditions could drastically vary. Thus, it is highly recommended for future studies to investigate the possibility of utilizing such progressed technologies in the reuse of Pb-212 in TAT. Similarly, atheromatous plaque imaging could be achieved by utilizing radionuclides. Through such non-invasive imaging, radionuclides are used to identify meaningful cellular or molecular processes [132]. Moreover, radionuclides can be used as tracers to detect leaks caused by corrosion, erosion, and wear in industrial processes. The endurance and reliability performance of machines, equipment, and industrial systems is significantly limited by corrosion, erosion, and wear. The absence of appropriate detection tools for the monitoring of such incidents can cause dangerous and unpredicted accidents. Thus, utilizing radionuclides as tracers is a viable method to monitor such processes [133].

8. Conclusion and future outlook

Nuclear wastewater management is increasingly becoming a concern with the widespread dependence on nuclear energy as a renewable source of energy. The large utilization of water in nuclear sectors raises questions as to how radioactive contaminated water is being handled. This review discusses nuclear wastewater's sources, classes, and environmental impacts and extensively reviews treatment technologies used for their decontamination. As discussed, because of the various sources of nuclear wastewater, initial classification is required to understand the properties and risks of all radioactive constituents present. Accordingly, suitable management approaches are chosen and taken to appropriately handle nuclear wastewater. Nuclear waste can be classified as EW, VSLW, VLLW, LLW, ILW, and HLW. The environmental footprint of nuclear wastewater results in negative implications on the environment in the case of improper disposal. After the Fukushima nuclear accident,

many research groups have studied the effects of radioactive leakages on the environment. Despite the very little quantitative analysis on the environmental effect of nuclear wastewater, there is evidence supporting its serious threats. The severity of released radioactive wastewater can only be assessed based on the classification of the released waste. Since most nuclear wastes can be categorized as EW or VLLW after concentration, they can be managed at conventional landfills or released to water reservoirs. The remaining concentrate, however, needs further processing as ILW [9]. The discharge of used cooling water, too, should be regarded as a major concern in the nuclear industry. Upon usage, cooling water is contaminated with radionuclides, making it unsafe to discharge directly into the environment. Improper treatment of such contaminated water could pose significant risks to the marine environment. In the case of not achieving the allowable discharge limits, waste is stored in high-integrity containers [134], also raising concerns with regard to the durability, lifespan, and cost-efficiency of such approaches given the large amounts of wastewater generated.

The following guidelines could be used when developing chemical, physical, electrochemical, or biological nuclear wastewater treatment technologies:

- Adsorption-based decontamination approaches should focus on the modification of natural adsorbents or the development of synthetic adsorbents as these have demonstrated higher decontamination rates, as compared to unmodified natural adsorbents. It was observed that the majority of adsorption studies took place at room temperature, leaving the effect of temperature unexplored. Similarly, the effect of radiation was not considered despite its significance. Thus, future work should consider the potential effects of real process conditions such as temperature and radiation in a way that mimics actual conditions in nuclear industries.
- Various technologies in membrane separation were explored for nuclear wastewater treatment. Among them, RO and MD had gained the greatest research attention. While RO can reject all kinds of ions, MD possesses the advantage of being low-energy consuming in the nuclear sector. NF membranes can reject multivalent ions, serving for the removal of some radionuclide ions. However, it is difficult to arrive at any conclusions with regard to their performance in nuclear wastewater with the currently available data. Future work on NF technology might prove its importance as a nuclear wastewater decontamination approach. Future directions should also focus on studying the effects of fouling, especially in RO processes, and explore more stable types of membranes instead of unstable polymeric membranes under radiation exposure.
- Facile chemical precipitation approaches were reported in literature as highly effective and economic. In those studies, it is assumed that chemical agent dosage is superior in its effect as compared to process conditions such as pH, temperature, and settling time. This assumption can be addressed in future studies to better assess chemical precipitation for nuclear wastewater decontamination.
- Evaporation was evidently used to achieve high volume reduction factors. However, this method suffers from high energy consumption when dealing with industrial-scale wastewater effluents. Regardless, research should continue to explore process modifications that decrease energy consumption such as MVR and solar-driven interfacial evaporation which aim at optimizing the processes' overall energy consumption.
- Electrochemical methods for nuclear wastewater treatment such as electrosorption, electrodialysis, and electrodeionization have been explored in literature. Unfortunately, this method encounters high electric resistance at low concentrations of radioactive ions. Thus, research on electrochemical methods in the nuclear sector must be conducted in more realistic settings to confirm their feasibility. This provides a good starting point for discussion and further research.
- Biological-based methods for nuclear wastewater decontamination use either biosorption or accumulation. In such techniques, limitations faced with adsorbents are re-encountered. As recommended above, future research should explore the effects of radiation and process conditions on the stability and performance of the utilized microorganisms.

Although various chemical, physical, and biological processes have been studied, this review demonstrates that, to date, adsorption and membrane separation are the two most researched technologies for nuclear wastewater treatment. It was also evident that hybrid technologies outperformed traditional ones due to their superior combined benefits at optimized process conditions. Future research on the development of an integrated hybrid radioactive wastewater treatment technology should address the following action items:

- Future investigations are necessary to validate the kinds of conclusions that are made from lab-scale studies. We believe that apart from analyzing lab-scale results, future research should consider more experimentation and scaled systems to better assess the feasibility of the studied treatment technologies in real nuclear wastewater. In other words, future work should shift towards the replication of bench-scale results in larger-scale systems.
- This review paper addresses the shift from nuclear wastewater treatment to R3-Reuse, Recover, and Resource Efficiency, so far lacking in scientific literature. Approaching wastewater management from a holistic perspective requires considering the reuse of water, recovery of valuables, and resource efficiency. Thus far, no discussions have been made with regard to the recovery of valuable radionuclides from nuclear wastewater. Recovered radionuclides can be reused in several applications, as discussed above. Therefore, this paper aims to highlight the significant need to identify new approaches to reuse beneficial radionuclides abundantly present in nuclear wastewater. The possibility of recovering radionuclides from nuclear wastewater warrants further investigations.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

All data generated or analysed during this study are included in this review article.

Acknowledgment

The authors acknowledge the Center for Membranes and Advanced Water Technology (CMAT) (Award No. RC2-2018-009) at Khalifa University for the support.

References

[1] V. Natarajan, M. Karunanidhi, B. Raja, A critical review on radioactive waste management through biological techniques, Recent Adv. Chem. Environ. Energy Eng. (2020) 29812–29823.
[2] G. Zakrzewska-Trznadel, Advances in membrane technologies for the treatment of liquid radioactive waste, Desalination 321 (2013) 119–130, https://doi.org/10.1016/j.desal.2013.02.022.
[3] B.M. Jun, H.K. Lee, S. Park, T.J. Kim, Purification of uranium-contaminated radioactive water by adsorption: a review on adsorbent materials, Sep. Purif. Technol. 278 (2022), 119675, https://doi.org/10.1016/j.seppur.2021.119675.
[4] D. Rana, T. Matsuura, M.A. Kassim, A.F. Ismail, Radioactive decontamination of water by membrane processes - a review, Desalination 321 (2013) 77–92, https://doi.org/10.1016/j.desal.2012.11.007.
[5] A.K. Pabby, B. Swain, N.L. Sonar, V.K. Mittal, T.P. Valsala, S. Ramsubramanian, D.B. Sathe, R.B. Bhatt, S. Pradhan, Radioactive waste processing using membranes: state of the art technology, challenges and perspectives, Sep. Purif. Rev. 51 (2022) 143–173, https://doi.org/10.1080/15422119.2021.1878221.

[6] S. Watanabe, H. Ogi, Y. Arai, H. Aihara, Y. Takahatake, A. Shibata, K. Nomura, Y. Kamiya, N. Asanuma, H. Matsuura, T. Kubota, N. Seko, T. Arai, T. Moriguchi, STRAD project for systematic treatments of radioactive liquid wastes generated in nuclear facilities, Prog. Nucl. Energy 117 (2019), 103090, https://doi.org/10.1016/j.pnucene.2019.103090.
[7] P. Vernon, Nuclear power, in: Power Gener. Technol, 2019, pp. 399–429, https://doi.org/10.1016/B978-0-08-102631-1.00017-1.
[8] V. Shestopalov, I. Shybetskyi, Ukraine: experience of radioactive waste (RAW) management and contaminated site clean-up, Radioact. Waste Manag. Contam. Site Clean-Up Process. Technol. Int. Exp. (2013) 386–414, https://doi.org/10.1533/9780857097446.2.386.
[9] R. Burcl, Radioactive waste (RAW) categories, characterization and processing route selection, Radioact. Waste Manag. Contam. Site Clean-Up Process. Technol. Int. Exp. (2013) 50–72, https://doi.org/10.1533/9780857097446.1.50.
[10] M.I. Hosan, Radioactive waste classification, management and environment, Eng. Int. 5 (2017) 53–62, https://doi.org/10.18034/ei.v5i2.178.
[11] J. Martínez, A. Peñalver, T. Baciu, M. Artigues, M. Danús, C. Aguilar, F. Borrull, Presence of artificial radionuclides in samples from potable water and wastewater treatment plants, J. Environ. Radioact. 192 (2018) 187–193, https://doi.org/10.1016/j.jenvrad.2018.06.024.
[12] S. Citation, Analysis of Cancer Risks in Populations Near Nuclear Facilities: Phase I, 2012, https://doi.org/10.17226/13388.
[13] T.Y. Kong, S. Kim, Y. Lee, J.K. Son, S.J. Maeng, Radioactive effluents released from korean nuclear power plants and the resulting radiation doses to members of the public, Nucl. Eng. Technol. 49 (2017) 1772–1777, https://doi.org/10.1016/j.net.2017.07.021.
[14] NEA, Effluent Release Options from Nuclear Installations, OECD Publishing, 2003.
[15] T. Mizutani, J. Koarashi, M. Takeishi, Monitoring of low-level radioactive liquid effluent in tokai reprocessing plant, J. Nucl. Sci. Technol. 46 (2009) 665–672, https://doi.org/10.1080/18811248.2007.9711573.
[16] E. Krawczyk, F. Piñero-García, M.A. Ferro-García, Discharges of nuclear medicine radioisotopes in spanish hospitals, J. Environ. Radioact. 116 (2013) 93–98, https://doi.org/10.1016/j.jenvrad.2012.08.011.
[17] M. Signoriello, M.R. Fornasier, M. de Denaro, F. Arfelli, B. Santoro, M. Severgnini, Assessment of total annual effective doses to representative person, for authorised and accidental releases from the nuclear medicine Department at Cattinara Hospital (Trieste, Italy), Phys. Med. 102 (2022) 88–95, https://doi.org/10.1016/j.ejmp.2022.09.001.
[18] R. Engelbrecht, Environmental Radioactivity Monitoring, Elsevier Inc, 2020, https://doi.org/10.1016/b978-0-12-814395-7.00001-5.
[19] S. Kim, T. Kim, H. Yang, Design of a low-Resolution Gamma-ray Spectrometer for monitoring radioactive levels of wastewater, Appl. Sci. 12 (2022), https://doi.org/10.3390/app12115613.
[20] Q. Guoxiu, Y. Xu, L. Wang, X. Zhang, W. Zhou, W. Li, Design of an on-line monitoring system for radioactive wastewater, J. Radioanal. Nucl. Chem. 314 (2017) 215–220, https://doi.org/10.1007/s10967-017-5356-1.
[21] A.L. Mosse, V.V. Savchin, V.V. Shilov, Testing and calculating a two-chamber plasma furnace for processing of radioactive wastes, J. Eng. Phys. Thermophys. 81 (2008) 598–606, https://doi.org/10.1007/s10891-008-0072-x.
[22] I. Ghiloufi, Simulation of radioelement volatility during the vitrification of radioactive wastes by arc plasma, J. Hazard. Mater. 163 (2009) 136–142, https://doi.org/10.1016/j.jhazmat.2008.06.110.
[23] E.S.P. Prado, F.S. Miranda, L.G. de Araujo, G. Petraconi, M.R. Baldan, Thermal plasma technology for radioactive waste treatment: a review, J. Radioanal. Nucl. Chem. 325 (2020) 331–342, https://doi.org/10.1007/s10967-020-07269-4.
[24] Y. Sun, N. Yuan, Y. Ge, T. Ye, Z. Yang, L. Zou, W. Ma, L. Lu, Adsorption behavior and mechanism of U(VI) onto phytic acid-modified Biochar/MoS2 heterojunction materials, Sep. Purif. Technol. 294 (2022), 121158, https://doi.org/10.1016/j.seppur.2022.121158.
[25] T. Xiong, Q. Li, J. Liao, Y. Zhang, W. Zhu, Highly enhanced adsorption performance to uranium(VI) by facile synthesized hydroxyapatite aerogel, J. Hazard. Mater. 423 (2022), 127184, https://doi.org/10.1016/j.jhazmat.2021.127184.
[26] M.B. Wu, S.C. Liu, J.Y. Fei, H. Ye, L.L. Ma, Natural cellulose-based microspheres decorated with amidoxime groups for decontamination of radioactive wastewater, Appl. Surf. Sci. 597 (2022), 153659, https://doi.org/10.1016/j.apsusc.2022.153659.
[27] Y. Ding, D. Liu, D. Luo, X. Sun, J. Mei, S. Wang, Z. Li, Rapid one-step preparation of a carboxymethyl chitosan gel with a novel crosslinker for efficient adsorption of Sr2+, Colloids Surf. A Physicochem. Eng. Asp. 641 (2022), 128576 https://doi.org/10.1016/j.colsurfa.2022.128576.
[28] J. Yang, K. Shi, F. Wu, J. Tong, Y. Su, T. Liu, Technetium-99 decontamination from radioactive wastewater by modified bentonite : batch, column experiment and mechanism investigation, Chem. Eng. J. 428 (2022), 131333, https://doi.org/10.1016/j.cej.2021.131333.
[29] Y.Ying Hu, C. Pan, S. Liu, X. Wang, X. Zheng, F. Hu, L. Xu, G. Xu, Y. Jian, X. Peng, Ultra-fast adsorption of radioactive-U(VI) and Cs(I) with high adsorption capacity towards CAA@MgAlFe spongy-like aerogel: mechanism and application feasibility study, J. Nucl. Mater. 559 (2022), https://doi.org/10.1016/j.jnucmat.2021.153463.
[30] Y. Ying Hu, C. Pan, X. Zheng, F. Hu, L. Xu, G. Xu, Y. Jian, X. Peng, Prediction and optimization of adsorption properties for Cs+on NiSiO@NiAlFe LDHs hollow spheres from aqueous solution: kinetics, isotherms, and BBD model, J. Hazard. Mater. 401 (2021), https://doi.org/10.1016/j.jhazmat.2020.123374.
[31] J. Yang, K. Shi, F. Wu, J. Tong, Y. Su, T. Liu, J. He, P. Mocilac, X. Hou, W. Wu, W. Shi, Technetium-99 decontamination from radioactive wastewater by modified bentonite: batch, column experiment and mechanism investigation, Chem. Eng. J. 428 (2022), 131333, https://doi.org/10.1016/j.cej.2021.131333.
[32] S.P. Mishra, S.S. Dubey, D. Tiwari, Ion-exchangers in radioactive waste management part XIV: removal behavior of hydrous titanium oxide and sodium titanate for Cs(I), J. Radioanal. Nucl. Chem. 261 (2004) 457–463, https://doi.org/10.1023/B:JRNC.0000034885.26277.bd.
[33] E.H. Borai, M.M.E. Breky, M.S. Sayed, M.M. Abo-Aly, Synthesis, characterization and application of titanium oxide nanocomposites for removal of radioactive cesium, cobalt and europium ions, J. Colloid Interface Sci. 450 (2015) 17–25, https://doi.org/10.1016/j.jcis.2015.02.062.
[34] Y. Tang, J. Zhou, J. Guo, X. Liao, B. Shi, Irradiation-stable hydrous titanium oxide-immobilized collagen fibers for uranium removal from radioactive wastewater, J. Environ. Manag. 283 (2021), 112001, https://doi.org/10.1016/j.jenvman.2021.112001.
[35] M.E. Mahmoud, E.A. Saad, A.M. El-Khatib, M.A. Soliman, E.A. Allam, Adsorptive removal of radioactive isotopes of cobalt and zinc from water and radioactive wastewater using TiO2/Ag2O nanoadsorbents, Prog. Nucl. Energy 106 (2018) 51–63, https://doi.org/10.1016/j.pnucene.2018.02.021.
[36] M. Zhang, P. Gu, S. Yan, L. Dong, G. Zhang, Na/Zn/Sn/S (NaZTS): quaternary metal sulfide nanosheets for efficient adsorption of radioactive strontium ions, Chem. Eng. J. 379 (2020), https://doi.org/10.1016/j.cej.2019.122227.
[37] M. Zhang, P. Gu, S. Yan, S. Pan, L. Dong, G. Zhang, A novel nanomaterial and its new application for efficient radioactive strontium removal from tap water: KZTS-NS metal sulfide adsorbent versus CTA-F-MF process, Chem. Eng. J. 391 (2020), https://doi.org/10.1016/j.cej.2019.123486.
[38] Z. Zhang, P. Gu, M. Zhang, S. Yan, L. Dong, G. Zhang, Synthesis of a robust layered metal sulfide for rapid and effective removal of Sr2+ from aqueous solutions, Chem. Eng. J. 372 (2019) 1205–1215, https://doi.org/10.1016/j.cej.2019.04.193.
[39] M. Zhang, P. Gu, Z. Zhang, J. Liu, L. Dong, G. Zhang, Effective, rapid and selective adsorption of radioactive Sr2+ from aqueous solution by a novel metal sulfide adsorbent, Chem. Eng. J. 351 (2018) 668–677, https://doi.org/10.1016/j.cej.2018.06.069.
[40] M. Zhang, P. Gu, S. Yan, Y. Liu, G. Zhang, Effective removal of radioactive cobalt from aqueous solution by a layered metal sulfide adsorbent: mechanism, adsorption performance, and practical application, Sep. Purif. Technol. 256 (2021), 117775, https://doi.org/10.1016/j.seppur.2020.117775.
[41] F. Liu, Z.F. Hu, M. Xiang, B. Hu, Magnetic amidoxime-functionalized MXenes for efficient adsorption and immobilization of U(VI) and Th(IV) from aqueous solution, Appl. Surf. Sci. 601 (2022), 154227, https://doi.org/10.1016/j.apsusc.2022.154227.
[42] B.M. Jun, M. Jang, C.M. Park, J. Han, Y. Yoon, Selective adsorption of Cs+ by MXene (Ti3C2Tx) from model low-level radioactive wastewater, Nucl. Eng. Technol. 52 (2020) 1201–1207, https://doi.org/10.1016/j.net.2019.11.020.
[43] Y. Sun, Y. Wei, J. Pei, H. Nan, Y. Wang, X. Cao, Y. Liu, Study on adsorption of U (VI) from MOF-derived phosphorylated porous carbons, J. Solid State Chem. 293 (2021), 121792, https://doi.org/10.1016/j.jssc.2020.121792.
[44] F.F. Li, W.R. Cui, W. Jiang, C.R. Zhang, R.P. Liang, J.D. Qiu, Stable sp2 carbon-conjugated covalent organic framework for detection and efficient adsorption of uranium from radioactive wastewater, J. Hazard. Mater. 392 (2020), 122333, https://doi.org/10.1016/j.jhazmat.2020.122333.
[45] Y. Ding, Q. Xian, Z. Jiang, L. Chen, T. Xiong, X. He, W. Yang, H. Dan, T. Duan, Immobilization of uranium in cristobalite ceramic through adsorption on mesoporous SBA-15 and further sintering process, J. Eur. Ceram. Soc. 40 (2020) 2113–2119, https://doi.org/10.1016/j.jeurceramsoc.2019.12.005.
[46] F. Liu, W. Huang, S. Wang, B. Hu, Investigation of adsorption properties and mechanism of uranium(VI) and europium(III) on magnetic amidoxime-functionalized MCM-41, Appl. Surf. Sci. 594 (2022), 153376, https://doi.org/10.1016/j.apsusc.2022.153376.
[47] C. Mao, H. Sang, Y. Chen, Y. Wei, Y. Wu, Journal of environmental chemical engineering layer-by-layer grafting synthesis of silicon-based zirconium organophosphate for the enhanced and selective adsorption of lanthanides, J. Environ. Chem. Eng. 10 (2022), 108608, https://doi.org/10.1016/j.jece.2022.108608.
[48] C. Dong, X. Deng, X. Guo, B. Wang, X. Ye, J. Fan, C. Zhu, F. Fan, B. Qing, Synthesis of potassium metal ferrocyanide/Al-MCM-41 with fast and selective adsorption of cesium, Colloids Surf. A Physicochem. Eng. Asp. 613 (2021), 126107, https://doi.org/10.1016/j.colsurfa.2020.126107.
[49] Y. Zhang, X. Nie, F. Dong, N. Pan, C. Ding, C. Liu, J. Wang, W. Cheng, M. Liu, H. He, S. Sun, X. Xia, Polyamine-modified polyacrylonitrile fibers for efficient removal of U(VI) from real fluorine-contained low-level radioactive wastewater, J. Water Process Eng. 45 (2022), 102452, https://doi.org/10.1016/j.jwpe.2021.102452.
[50] D. Wen, Z. Dong, Y. Ao, M. Kangjun Xie, L.Zhao Zhai, Aminotriazole isomers modified cellulose microspheres for selective adsorption of U(VI): performance and mechanism investigation, Carbohydr. Polym. 257 (2021), 117666, https://doi.org/10.1016/j.carbpol.2021.117666.
[51] E. Karabayir, O.S. Taskin, A. Ozdemir, B.F. Senkal, Effective removal of 137Cs ions from radioactive wastewater by melamine-styrene based polymer (MSP), Appl. Radiat. Isot. 176 (2021), 109912, https://doi.org/10.1016/j.apradiso.2021.109912.
[52] I.-T. Hwang, D.-S. Han, J.-Y. Sohn, J. Shin, J.-H. Choi, C.-H. Jung, Preparation and cesium adsorption behavior of prussian blue-based polypropylene nonwoven fabric by surfactant-assisted aqueous preirradiation graft polymerization, Radiat.

Phys. Chem. 199 (2022), 110356, https://doi.org/10.1016/j.radphyschem.2022.110356.
[53] Y. Huang, H. Zheng, H. Li, Z. Zhang, C. Zhao, Q. Gou, Y. Liu, Highly effective and selective adsorption of thorium(IV) from aqueous solution using mesoporous graphite carbon nitride prepared by sol–gel template method, Chem. Eng. J. 410 (2021), 128321, https://doi.org/10.1016/j.cej.2020.128321.
[54] X. Tao, Y. Fang, Preparation of amidoxime modified calixarene fiber for highly efficient adsorption of uranium (VI), Sep. Purif. Technol. 303 (2022), 122257, https://doi.org/10.1016/j.seppur.2022.122257.
[55] Y. Tian, L. Liu, F. Ma, X. Zhu, H. Dong, C. Zhang, F. Zhao, Synthesis of phosphorylated hyper-cross-linked polymers and their efficient uranium adsorption in water, J. Hazard. Mater. 419 (2021), 126538, https://doi.org/10.1016/j.jhazmat.2021.126538.
[56] Y. Wang, Z. Lin, Q. Liu, J. Zhu, J. Liu, J. Yu, R. Chen, P. Liu, J. Wang, Ultra-high mechanical property and multi-layer porous structure of amidoximation ethylene-acrylic acid copolymer balls for efficient and selective uranium adsorption from radioactive wastewater, Chemosphere 280 (2021), 130722, https://doi.org/10.1016/j.chemosphere.2021.130722.
[57] J. Yin, S. Yang, W. He, T. Zhao, C. Li, D. Hua, Biogene-derived aerogels for simultaneously selective adsorption of uranium(VI) and strontium(II) by co-imprinting method, Sep. Purif. Technol. 271 (2021), 118849, https://doi.org/10.1016/j.seppur.2021.118849.
[58] D. Wang, H. Chen, J. Zhang, J. Li, Easily synthesized mesoporous aluminum phosphate for the enhanced adsorption performance of U(VI) from aqueous solution, J. Hazard. Mater. 432 (2022), 128675, https://doi.org/10.1016/j.jhazmat.2022.128675.
[59] L. Chen, H. Wang, X. Cao, Y. Feng, Z. Zhang, Y. Wang, Y. Liu, Effects of different phosphorus sources on the adsorption of U(VI) by Zr(IV) organophosphate hybrids, J. Solid State Chem. 302 (2021), 122434, https://doi.org/10.1016/j.jssc.2021.122434.
[60] H. Liao, Y. Li, H. Li, B. Li, Y. Zhou, D. Liu, X. Wang, Efficiency and mechanism of amidoxime-modified X-type zeolite (AO-XZ) for Cs+ adsorption, Chem. Phys. Lett. 741 (2020), https://doi.org/10.1016/j.cplett.2019.137084.
[61] W. Han, Y. Huang, M. Su, H. Liu, C. Shen, Y. Zhou, T. Ou, D. Chen, Highly selective adsorption and lattice process of cesium by cubic cyanide-based functional materials, Environ. Res. 214 (2022), 114085, https://doi.org/10.1016/j.envres.2022.114085.
[62] Z. Dong, Z. Li, D. Zeng, Z. Cheng, Y. Wang, Y. Dai, X. Cao, Y. Wang, Z. Zhang, Y. Liu, Highly selective adsorption of radioactive cesium by Novel Calix [4] biscrown-6 functionalized millimetre-sized, Sep. Purif. Technol. (2022), 122255, https://doi.org/10.1016/j.seppur.2022.122255.
[63] X. Tao, Y. Fang, Preparation of modified calixarene fiber for highly efficient adsorption of uranium (VI), Sep. Purif. Technol. (2022), 122257, https://doi.org/10.1016/j.seppur.2022.122257.
[64] W. Mu, B. Chen, X. Li, H. Wei, Y. Yang, S. Peng, Preparation of novel zirconium phosphonates for adsorption of lutetium, Nucl. Anal. 1 (2022), 100010, https://doi.org/10.1016/j.nucana.2022.100010.
[65] R. Zhou, Z. Zhang, Z. Cheng, Y. Dai, Y. Liu, Y. Wang, Y. Xie, Z. Zhang, X. Cao, Y. Liu, Selective adsorption of uranium from aqueous solution by mesoporous polymer-carbon composites containing phosphate groups, Environ. Nanotechnology, Monit. Manag. 15 (2021), 100441, https://doi.org/10.1016/j.enmm.2021.100441.
[66] D. Wang, Y. Xu, D. Xiao, Q. Qiao, P. Yin, Z. Yang, J. Li, W. Winchester, Z. Wang, T. Hayat, Ultra-thin iron phosphate nanosheets for high efficient U(VI) adsorption, J. Hazard. Mater. 371 (2019) 83–93, https://doi.org/10.1016/j.jhazmat.2019.02.091.
[67] A.E. Osmanlioglu, Decontamination of radioactive wastewater by two-staged chemical precipitation, Nucl. Eng. Technol. 50 (2018) 886–889, https://doi.org/10.1016/j.net.2018.04.009.
[68] A.V. Gusa, A. Tomani, Z. Zhang, R.D. Vidic, Sulfate precipitation in produced water from Marcellus shale for the control of naturally occurring radioactive material, Water Res. 177 (2020), 115765, https://doi.org/10.1016/j.watres.2020.115765.
[69] B. Ouyang, D.J. Renock, M.A. Ajemigbitse, K. Van, N.R. Warner, D. Landis, X. Feng, Radium in hydraulic fracturing wastewater : its treatment by sulfate co-precipitation †, Environ Sci Process Impacts (2019) 339–351, https://doi.org/10.1039/c8em00311d.
[70] C. Jiao, G. Wang, J. Wang, Y. Gao, H. Hou, M. Zhang, Y. Li, Effects of coprecipitation conditions on - cs + removal, coprecipitate compositions and coprecipitate particle-size distribution in nickel potassium ferrocyanide systems, J. Radioanal. Nucl. Chem. 330 (2021) 293–303, https://doi.org/10.1007/s10967-021-07961-z.
[71] C. Hou, W. Lin, L. Yang, H. Zhang, Experimental performance evaluation and model-based optimal design of a mechanical vapour recompression system for radioactive wastewater treatment, Energy Convers. Manag. 252 (2022), 115087, https://doi.org/10.1016/j.enconman.2021.115087.
[72] K. Yu, P. Shao, P. Meng, T. Chen, J. Lei, X. Yu, R. He, F. Yang, W. Zhu, T. Duan, Superhydrophilic and highly elastic monolithic sponge for efficient solar-driven radioactive wastewater treatment under one sun, J. Hazard. Mater. 392 (2020), 122350, https://doi.org/10.1016/j.jhazmat.2020.122350.
[73] S. Yu, X. Zhang, F. Li, X. Zhao, Poly(vinyl pyrrolidone) modified poly(vinylidene fluoride) ultrafiltration membrane via a two-step surface grafting for radioactive wastewater treatment, Sep. Purif. Technol. 194 (2018) 404–409, https://doi.org/10.1016/j.seppur.2017.10.051.
[74] B. Chen, D. Chen, X. Zhao, Radioactive wastewater treatment with modified aromatic polyamide reverse osmosis membranes via quaternary ammonium cation grafting, Sep. Purif. Technol. 252 (2020), 117378, https://doi.org/10.1016/j.seppur.2020.117378.
[75] B. Chen, S. Yu, X. Zhao, The influence of membrane surface properties on the radionuclide mass transfer process in reverse osmosis, Sep. Purif. Technol. 252 (2020), 117455, https://doi.org/10.1016/j.seppur.2020.117455.
[76] B. Chen, S. Yu, X. Zhao, The influence of RO membrane surface properties on surfactant fouling in radioactive wastewater treatment, Process Saf. Environ. Prot. 149 (2021) 858–865, https://doi.org/10.1016/j.psep.2021.03.006.
[77] J. Li, S. Zhuang, L. Wang, J. Wang, Treatment of radioactive wastewater from high-temperature gas-cooled reactor by membrane system, Nucl. Technol. 203 (2018) 101–107, https://doi.org/10.1080/00295450.2018.1432838.
[78] L. Chen, X. Bian, X. Lu, Removal of strontium from simulated low-level radioactive wastewater by nanofiltration, Water Sci. Technol. 78 (2018) 1733–1740, https://doi.org/10.2166/wst.2018.455.
[79] X. Liu, J. Wu, J. Wang, Removal of Cs(I) from simulated radioactive wastewater by three forward osmosis membranes, Chem. Eng. J. 344 (2018) 353–362, https://doi.org/10.1016/j.cej.2018.03.046.
[80] X. Liu, J. Wu, L. An Hou, J. Wang, Removal of Co, Sr and Cs ions from simulated radioactive wastewater by forward osmosis, Chemosphere 232 (2019) 87–95, https://doi.org/10.1016/j.chemosphere.2019.05.210.
[81] X. Liu, J. Wu, J. Wang, Removal of nuclides and boric acid from simulated radioactive wastewater by forward osmosis, Prog. Nucl. Energy 114 (2019) 155–163, https://doi.org/10.1016/j.pnucene.2019.03.014.
[82] B. Chen, S. Yu, X. Zhao, The separation of radionuclides and silicon from boron-containing radioactive wastewater with modified reverse osmosis membranes, Process Saf. Environ. Prot. 146 (2021) 639–646, https://doi.org/10.1016/j.psep.2020.11.023.
[83] X. Liu, J. Wu, L. An Hou, J. Wang, Fouling and cleaning protocols for forward osmosis membrane used for radioactive wastewater treatment, Nucl. Eng. Technol. 52 (2020) 581–588, https://doi.org/10.1016/j.net.2019.08.007.
[84] W. Xia, Y. Cheng-long, Z. Xuan, Treatment of simulated radioactive wastewater by direct contact membrane distillation, J. Nucl. Radiochem. 42 (2020).
[85] M. Essalhi, N. Ismail, S. Tesfalidet, J. Pan, Q. Wang, Z. Cui, M.C. García-Payo, M. Khayet, J.P. Mikkola, S. Sarmad, D. Bouyer, Y. Zhao, B. Li, C. André Ohlin, N. Tavajohi, Polyvinylidene fluoride membrane formation using carbon dioxide as a non-solvent additive for nuclear wastewater decontamination, Chem. Eng. J. 446 (2022), https://doi.org/10.1016/j.cej.2022.137300.
[86] X. Nie, X. Hu, C. Liu, X. Xia, F. Dong, Decontamination of uranium contained low-level radioactive wastewater from UO2 fuel element industry with vacuum membrane distillation, Desalination 516 (2021), 115226, https://doi.org/10.1016/j.desal.2021.115226.
[87] X. Jia, L. Lan, X. Zhang, T. Wang, Y. Wang, C. Ye, J. Lin, Pilot-scale vacuum membrane distillation for decontamination of simulated radioactive wastewater: system design and performance evaluation, Sep. Purif. Technol. 275 (2021), 119129, https://doi.org/10.1016/j.seppur.2021.119129.
[88] F. Jia, Y. Yin, J. Wang, Removal of cobalt ions from simulated radioactive wastewater by vacuum membrane distillation, Prog. Nucl. Energy 103 (2018) 20–27, https://doi.org/10.1016/j.pnucene.2017.11.008.
[89] S. Li, Z. He, D. Xiao, L. Guo, Study on the treatment of radioactive wastewater by non-contact membrane distillation, Sep. Purif. Technol. 290 (2022), 120766, https://doi.org/10.1016/j.seppur.2022.120766.
[90] C.A. Martínez-Huitle, S. Ferro, Electrochemical oxidation of organic pollutants for the wastewater treatment: direct and indirect processes, Chem. Soc. Rev. 35 (2006) 1324–1340, https://doi.org/10.1039/b517632h.
[91] H. Yu, L. Zhou, Z. Li, Y. Liu, X. Ao, J. Ouyang, Z. Le, Z. Liu, A.A. Adesina, Electrodeposited polypyrrole/biomass-derived carbon composite electrodes with high hybrid capacitance and hierarchical porous structure for enhancing U(VI) electrosorption from aqueous solution, Sep. Purif. Technol. 302 (2022), 122169, https://doi.org/10.1016/j.seppur.2022.122169.
[92] S. Xiang, H. Mao, W. Geng, Y. Xu, H. Zhou, Selective removal of Sr(II) from saliferous radioactive wastewater by capacitive deionization, J. Hazard. Mater. 431 (2022), 128591, https://doi.org/10.1016/j.jhazmat.2022.128591.
[93] J. Zhou, X. Zhang, Y. Zhang, D. Wang, H. Zhou, J. Li, Effective inspissation of uranium(VI) from radioactive wastewater using flow electrode capacitive deionization, Sep. Purif. Technol. 283 (2022), 120172, https://doi.org/10.1016/j.seppur.2021.120172.
[94] J. Zhou, H. Zhou, Y. Zhang, J. Wu, H. Zhang, G. Wang, J. Li, Pseudocapacitive deionization of uranium(VI) with WO3/C electrode, Chem. Eng. J. 398 (2020), 125460, https://doi.org/10.1016/j.cej.2020.125460.
[95] X. Yang, L. Jiang, S. Peng, L. Zhang, G. Huang, Removal of cesium from liquid radioactive waste by in situ electrochemical synthesis of cesium zinc ferrocyanide, Colloids Surf. A Physicochem. Eng. Asp. 626 (2021), 127050, https://doi.org/10.1016/j.colsurfa.2021.127050.
[96] S. Yan Lv, M. Li, X. Yan Wu, X. Wen Zhang, Y. Long Hua, L. Bi, Q. Fang, T. Cai, A non-polluting method for rapidly purifying uranium-containing wastewater and efficiently recovering uranium through electrochemical mineralization and oxidative roasting, J. Hazard. Mater. 416 (2021), https://doi.org/10.1016/j.jhazmat.2021.125885.
[97] H. Yu, Y. Dai, L. Zhou, J. Ouyang, X. Tang, Z. Liu, A.A. Adesina, Selective biosorption of U(VI) from aqueous solution by ion-imprinted honeycomb-like chitosan/kaolin clay composite foams, Int. J. Biol. Macromol. 206 (2022) 409–421, https://doi.org/10.1016/j.ijbiomac.2022.02.168.
[98] H. Ding, X. Luo, X. Zhang, H. Yang, Alginate-immobilized Aspergillus niger: characterization and biosorption removal of thorium ions from radioactive wastewater, Colloids Surf. A Physicochem. Eng. Asp. 562 (2019) 186–195, https://doi.org/10.1016/j.colsurfa.2018.11.032.

[99] H. Ding, X. Zhang, H. Yang, Y. Zhang, X. Luo, Biosorption of U(VI) by active and inactive aspergillus Niger: equilibrium, kinetic, thermodynamic and mechanistic analyses, J. Radioanal. Nucl. Chem. 319 (2019) 1261–1275, https://doi.org/10.1007/s10967-019-06420-0.
[100] J. Liang, L. Liu, W. Song, Enhancement of U(VI) biosorption by trichoderma harzianum mutant obtained by a cold atmospheric plasma jet, J. Radioanal. Nucl. Chem. 327 (2021) 1325–1333, https://doi.org/10.1007/s10967-021-07615-0.
[101] Q. Zhang, S. Zhuang, J. Wang, Biosorptive removal of cobalt(II) from aqueous solutions using magnetic cyanoethyl chitosan beads, J. Environ. Chem. Eng. 8 (2020), 104531, https://doi.org/10.1016/j.jece.2020.104531.
[102] Y. Yuan, X. Liao, Z. Fang, N. Liu, F. Qiu, D. Huang, B. Wang, Y. Zhou, Study of the performance of a bloom-forming cyanobacterium (Microcystis aeruginosa) on the biosorption of uranium, J. Radioanal. Nucl. Chem. 331 (2022) 3183–3194, https://doi.org/10.1007/s10967-022-08384-0.
[103] T.Y. Kim, S.H. Lee, S.Y. Lee, Two newly identified haematococcus strains efficiently accumulated radioactive cesium over higher astaxanthin production, Environ. Res. 199 (2021), 111301, https://doi.org/10.1016/j.envres.2021.111301.
[104] H.M. Saleh, T.A. Bayoumi, H.H. Mahmoud, R.F. Aglan, Uptake of cesium and cobalt radionuclides from simulated radioactive wastewater by Ludwigia stolonifera aquatic plant, Nucl. Eng. Des. 315 (2017) 194–199, https://doi.org/10.1016/j.nucengdes.2017.02.018.
[105] I. Kim, H.M. Yang, C.W. Park, I.H. Yoon, B.K. Seo, E.K. Kim, B.G. Ryu, Removal of radioactive cesium from an aqueous solution via bioaccumulation by microalgae and magnetic separation, Sci. Rep. 9 (2019) 3–10, https://doi.org/10.1038/s41598-019-46586-x.
[106] H.M. Saleh, H.R. Moussa, H.H. Mahmoud, F.A. El-Saied, M. Dawoud, R.S. Abdel Wahed, Potential of the submerged plant Myriophyllum spicatum for treatment of aquatic environments contaminated with stable or radioactive cobalt and cesium, Prog. Nucl. Energy 118 (2020), 103147, https://doi.org/10.1016/j.pnucene.2019.103147.
[107] R. Cheng, M. Kang, S. Zhuang, L. Shi, X. Zheng, J. Wang, Adsorption of Sr(II) from water by mercerized bacterial cellulose membrane modified with EDTA, J. Hazard. Mater. 364 (2019) 645–653, https://doi.org/10.1016/j.jhazmat.2018.10.083.
[108] S. Bolisetty, N.M. Coray, A. Palika, G.A. Prenosil, R. Mezzenga, Amyloid hybrid membranes for removal of clinical and nuclear radioactive wastewater, Environ. Sci. Water Res. Technol. 6 (2020) 3249–3254, https://doi.org/10.1039/d0ew00693a.
[109] Y. Kim, H.H. Eom, D. Kim, D. Harbottle, J.W. Lee, Adsorptive removal of cesium by electrospun nanofibers embedded with potassium copper hexacyanoferrate, Sep. Purif. Technol. 255 (2021), 117745, https://doi.org/10.1016/j.seppur.2020.117745.
[110] X. Zhang, Y. Liu, Integrated forward osmosis-adsorption process for strontium-containing water treatment: pre-concentration and solidification, J. Hazard. Mater. 414 (2021), 125518, https://doi.org/10.1016/j.jhazmat.2021.125518.
[111] W. Luo, J. Chen, H. Lin, X. Ye, Biomass base membrane with phenol hydroxy-amino group for ultraselective adsorption of radioactive Co(II) in trace concentration, Sep. Purif. Technol. 272 (2021), 118878, https://doi.org/10.1016/j.seppur.2021.118878.
[112] S. Ding, L. Zhang, Y. Li, L. An Hou, Fabrication of a novel polyvinylidene fluoride membrane via binding SiO2 nanoparticles and a copper ferrocyanide layer onto a membrane surface for selective removal of cesium, J. Hazard. Mater. 368 (2019) 292–299, https://doi.org/10.1016/j.jhazmat.2019.01.065.
[113] S. Xuan, B. Zhang, L. Xiao, G. Li, Y. Zhang, Y. Zhang, J. Li, Facile carboxylation of natural eggshell membrane for highly selective uranium (VI) adsorption from radioactive wastewater, Environ. Sci. Pollut. Res. 28 (2021) 45134–45143, https://doi.org/10.1007/s11356-021-13820-x.
[114] W. Luo, H. Xu, Y. Wang, X. Ye, C. Luo, X. Liao, X. Huang, B. Shi, Radionuclide tolerance mechanism of plants for ultraselective enrichment of low content of thorium with exceptional selectivity coefficient, J. Hazard. Mater. 380 (2019), 120893, https://doi.org/10.1016/j.jhazmat.2019.120893.
[115] X. Long, Y.S. Chen, Q. Zheng, X.X. Xie, H. Tang, L.P. Jiang, J.T. Jiang, J.H. Qiu, Removal of iodine from aqueous solution by PVDF/ZIF-8 nanocomposite membranes, Sep. Purif. Technol. 238 (2020), 116488, https://doi.org/10.1016/j.seppur.2019.116488.
[116] Y. Wang, J. Long, W. Xu, H. Luo, J. Liu, Y. Zhang, J. Li, X. Luo, Removal of uranium(VI) from simulated wastewater by a novel porous membrane based on crosslinked chitosan, UiO-66-NH2 and polyvinyl alcohol, J. Radioanal. Nucl. Chem. 328 (2021) 397–410, https://doi.org/10.1007/s10967-021-07649-4.
[117] D. Park, J.H. Kim, H.J. Kim, D. Lee, D.S. Lee, D.S. Yoon, K.S. Hwang, Multiplexed femtomolar detection of Alzheimer's disease biomarkers in biofluids using a reduced graphene oxide field-effect transistor, Biosens. Bioelectron. 167 (2020), 112505, https://doi.org/10.1016/j.bios.2020.112505.
[118] J.H. Park, H. Kim, M. Kim, J.M. Lim, J. Ryu, S. Kim, Sequential removal of radioactive cs by electrochemical adsorption and desorption reaction using core-shell structured carbon nanofiber–Prussian blue composites, Chem. Eng. J. 399 (2020), 125817, https://doi.org/10.1016/j.cej.2020.125817.
[119] C. Jiao, H. Wang, Y. Li, M. Zhang, Y. Gao, M. He, Treatment of simulated radioactive wastewater using reverse osmosis and membrane distillation, Nucl. Technol. 00 (2022) 1–9, https://doi.org/10.1080/00295450.2022.2081483.
[120] J.W. Suh, S.Y. Sohn, B.K. Lee, Patent clustering and network analyses to explore nuclear waste management technologies, Energy Policy 146 (2020), 111794, https://doi.org/10.1016/j.enpol.2020.111794.
[121] F. Tibi, A. Charfi, J. Cho, J. Kim, Fabrication of polymeric membranes for membrane distillation process and application for wastewater treatment: critical review, Process Saf. Environ. Prot. 141 (2020) 190–201, https://doi.org/10.1016/j.psep.2020.05.026.
[122] J.M. Arnal, M. Sancho, B. García-Fayos, G. Verdú, C. Serrano, J.T. Ruiz-Martínez, Declassification of radioactive water from a pool type reactor after nuclear facility dismantling, Radiat. Phys. Chem. 138 (2017) 72–74, https://doi.org/10.1016/j.radphyschem.2017.03.027.
[123] M. Živanović, I. Vukanac, J. Samac, V. Antić, M. Jeremić, D. Janković, A. Kandić, C.S. Vargas, L. Struelens, O. Ciraj-Bjelac, Intercomparison and performance assessment of radionuclide calibrators used in nuclear medicine departments in Serbia, Appl. Radiat. Isot. 179 (2022), https://doi.org/10.1016/j.apradiso.2021.110013.
[124] S.J. Adelstein, F.J. Manning, Chapter 3: reactor-produced radionuclides, in: Isot. Med. Life Sci, 1995, https://doi.org/10.17226/4818.
[125] F.D.C. Guerra Liberal, J.M. O'Sullivan, S.J. McMahon, K.M. Prise, Targeted alpha therapy: current clinical applications, Cancer Biother. Radiopharm. 35 (2020) 404–417, https://doi.org/10.1089/cbr.2020.3576.
[126] K.V. Kokov, B.V. Egorova, M.N. German, I.D. Klabukov, M.E. Krasheninnikov, A. A. Larkin-Kondrov, K.A. Makoveeva, M.V. Ovchinnikov, M.V. Sidorova, D. Y. Chuvilin, 212 pb: production approaches and targeted therapy applications, Pharmaceutics 14 (2022) 1–21, https://doi.org/10.3390/pharmaceutics14010189.
[127] C.L. Corkhill, J.W. Bridge, P. Hillel, L.J. Gardner, S.A. Banwart, N.C. Hyatt, Technetium-99m transport and immobilisation in porous media: development of a novel nuclear imaging technique, Mater. Res. Soc. Symp. Proc. 1518 (2013) 123–129, https://doi.org/10.1557/opl.2013.111.
[128] I.M. El-Naggar, S.A. Ahmed, N. Shehata, E.S. Sheneshen, M. Fathy, A. Shehata, A novel approach for the removal of lead (II) ion from wastewater using Kaolinite/Smectite natural composite adsorbent, Appl Water Sci 9 (2019) 1–13, https://doi.org/10.1007/s13201-018-0845-0.
[129] Z. Samavati, A. Samavati, P. Sean, A comprehensive review of recent advances in nanofiltration membranes for heavy metal removal from wastewater, Chem. Eng. Res. Des. 189 (2023) 530–571, https://doi.org/10.1016/j.cherd.2022.11.042.
[130] F. Zhao, H. Xiang, X. Min, C. Tang, Recent advances in membrane filtration for heavy metal removal from wastewater : a mini review, J. Water Process Eng. 49 (2022), https://doi.org/10.1016/j.jwpe.2022.103023.
[131] V. Kumar, S.K. Dwivedi, S. Oh, A critical review on lead removal from industrial wastewater : recent advances and future outlook, J. Water Process Eng. 45 (2022), 102518, https://doi.org/10.1016/j.jwpe.2021.102518.
[132] J.R. Davies, J.H.F. Rudd, P.L. Weissberg, J. Narula, Radionuclide imaging for the detection of inflammation in vulnerable plaques, J. Am. Coll. Cardiol. 47 (2006), https://doi.org/10.1016/j.jacc.2005.11.049.
[133] International Atomic Energy Agency, in: Radiotracer Technologies for Wear, Erosion and Corrosion Management, 2020, p. 98.
[134] D. Englehardt, Pozzolanic filtration/solidification of radionuclides in nuclear reactor cooling water, Waste Manag. 15 (1996) 585–592.